RESEARCH STUDIES ON ELECTROMAGNETICALLY INDUCED TRANSPARENCY

FINAL PROGRESS REPORT

(8 JANUARY 1998 - 7 JANUARY 2001)

S. E. HARRIS

U.S. ARMY RESEARCH OFFICE

GRANT DAAG-55-98-1-0031

Edward L. Ginzton Laboratory
Stanford University
Stanford, California

APRIL 2001

MASTER COPY KEEP THIS COPY FOR REPRODUCTION PURPOSES

REPORT DOCUMENTATION PAGE

Form Approved
OMB NO. 0704-0188

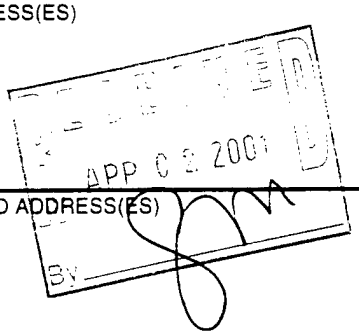
1. AGENCY USE ONLY (Leave blank)	2. REPORT DATE	3. REPORT TYPE AND DATES COVERED
	April 2001	8 January 1998 - 7 January 2001 FINAL

4. TITLE AND SUBTITLE
"Research Studies on Electromagnetically Induced Transparency"

5. FUNDING NUMBERS
DAAG55-98-1-0031

6. AUTHOR(S)
S. E. Harris

7. PERFORMING ORGANIZATION NAMES(S) AND ADDRESS(ES)
Edward L. Ginzton Laboratory
Stanford University
Stanford, CA 94305-4085



8. PERFORMING ORGANIZATION REPORT NUMBER

9. SPONSORING / MONITORING AGENCY NAME(S) AND ADDRESS(ES)
U.S. Army Research Office
P.O. Box 12211
Research Triangle Park, NC 27709-2211

10. SPONSORING / MONITORING AGENCY REPORT NUMBER
37331-PH
.24

11. SUPPLEMENTARY NOTES
The views, opinions and/or findings contained in this report are those of the author(s) and should not be construed as an official Department of the Army position, policy or decision, unless so designated by other documentation.

12a. DISTRIBUTION / AVAILABILITY STATEMENT
Approved for public release; distribution unlimited.

12 b. DISTRIBUTION CODE

13. ABSTRACT (Maximum 200 words)

We describe experimental and theoretical advances in the use of electromagnetically induced transparency (EIT) for new types of optical devices. Highlights of our work over the cited three-year period include: (1) The first demonstration that light may be made to propagate at ultra-slow group velocities. (2) The suggestion that slow light may lead to a class of nonlinear optical processes which are sufficiently efficient that they may occur at the single-photon level. (3) The experimental demonstration of high-efficiency nonlinear optics in gas phase. In particular, we obtain complete conversion from a 233-nm optical field to a 186-nm field. (4) We have suggested and experimentally demonstrated a new type of EIT-based Raman molecular process. We have demonstrated a source which produces mutually-coherent, equidistant sidebands covering about 50,000 wavenumbers of spectral bandwidth and ranging from 2.94 microns to 195 nm in wavelength. (5) We have shown that we may phase several of these sidebands to produce amplitude- or frequency-modulated light with a modulation frequency which is equal to the fundamental vibrational frequency of molecular deuterium (approximately 90 THz). This experiment is a first step toward the synthesis of subfemtosecond pulses with prescribed temporal shape.

14. SUBJECT TERMS
Electromagnetically induced transparency, slow light, low light level nonlinear optics, subfemtosecond pulses

15. NUMBER IF PAGES
68

16. PRICE CODE

17. SECURITY CLASSIFICATION OR REPORT	18. SECURITY CLASSIFICATION OF THIS PAGE	19. SECURITY CLASSIFICATION OF ABSTRACT	20. LIMITATION OF ABSTRACT
UNCLASSIFIED	UNCLASSIFIED	UNCLASSIFIED	UL

TABLE OF CONTENTS

I. FORWARD

The central theme of our work over the past three years has been the use of electromagnetically induced transparency (EIT) for new types of nonlinear interactions and processes. The last several years have been unusually productive. In a February 1999 *Nature* article which received world-wide attention, Lene Hau, her students, and the author of this report, Steve Harris, showed that, by using EIT in an ultra-cold gas of Na atoms, an optical pulse could be caused to travel 10^7 times slower than the speed of light in vacuum. In a June 1991 *Physical Review Letter*, Harris and Hau showed how this slow group velocity could result in nonlinear coefficients which are sufficiently large that it may soon be possible to do nonlinear optics at picowatt power levels. In a May 1999 *Optics Letter*, the technique of nonlinear optics at maximum coherence was used to demonstrate efficient gas phase frequency conversion to the vacuum ultraviolet. In particular, a 233-nm laser beam was converted to 186 nm at approximately unity photon efficiency. In follow-up work, this same technique was extended to a system where all frequencies are resonant. Here, a conversion efficiency of about 30% was obtained at laser power densities of about 10^5 W/cm^2 and at atom densities of only 10^{14} atoms/cm^3. In another area of substantial progress, Harris and Sokolov have suggested a new technique for producing subfemtosecond and even single-cycle optical pulses. The idea is to use two laser beams whose frequency difference is slightly offset from a molecular transition. This produces a single, molecular, superposition state which, in the manner of EIT, may be driven at maximum coherence. In recent experimental work, we have shown the collinear generation of equidistant

sidebands covering about 50,000 cm^{-1} of spectral bandwidth and ranging from 2.94 μ in the infrared to 195 nm in the ultraviolet. This work is the first demonstration of a nonlinear optical process at maximum coherence in a molecular system. Because these sidbands comprise an FM signal, group velocity dispersion may be used to temporally compress this waveform to a train of subfemtosecond pulses.

II. STATEMENT OF PROBLEM STUDIED

We study electromagnetically induced transparency (EIT), both in its own right and as a vehicle to make new types of nonlinear optical devices. Examples of such devices include subfemtosecond and single-cycle pulse generators and nonlinear optical converters which may function at the single-photon level.

III. SUMMARY OF MOST IMPORTANT RESULTS

To show the overall evolution of this work the Summary of Important Results includes the main contributions of our earlier contracts in this area.

(1) Our work in this field began with the paper: S. E. Harris, J. E. Field, and A. Imamoglu, "Nonlinear Optical Processes Using Electromagnetically Induced Transparency," *Phys. Rev. Lett.* **64**, 1107-1110 (March 1990). This work also introduced the expression "electromagnetically induced transparency."

(2) The first observation of EIT was reported in: K.-J. Boller, A. Imamoglu, and S. E. Harris, "Observation of Electromagnetically Induced Transparency," *Phys. Rev. Lett.* **66**, 2593-2596 (May 1991).

(3) The concept of matched pulses and the normal modes of EIT were introduced in: S. E. Harris, "Electromagnetically Induced Transparency with Matched Pulses," *Phys. Rev. Lett.* **70**, 552-555 (February 1993) and S. E. Harris, "Normal Modes for Electromagnetically Induced Transparency," *Phys. Rev. Lett.* **72**, 52-55 (January 1994).

(4) In March of 1995 we showed how EIT would cause well-shaped pulses to propagate at a group velocity as slow as c/165: A. Kasapi, M. Jain, G. Y. Yin, and S. E. Harris, "Electromagnetically Induced Transparency: Propagation Dynamics," *Phys. Rev. Lett.* **74**, 1447-2450 (March 1995).

(5) The observation that EIT would also eliminate self-focusing and defocusing was described in: M. Jain, A. J. Merriam, A. Kasapi, G. Y. Yin, and S. E. Harris, "Elimination of Optical Self-Focusing by Population Trapping," *Phys. Rev. Lett.* **75**, 4385-4388 (December 1995).

(6) The idea of nonlinear optics at maximum coherence was introduced and experimentally demonstrated in the paper: M. Jain, H. Xia, G. Y. Yin, A. J. Merriam, and S. E. Harris, "Efficient Nonlinear Frequency Conversion with Maximal Atomic Coherence," *Phys. Rev. Lett.* **77**, 4326-4329 (November 1996).

(7) The suggestion for subfemtosecond pulse generation by molecular modulation was made in: S. E. Harris and A. V. Sokolov, "Subfemtosecond Pulse Generation by Molecular Modulation," *Phys. Rev. Lett.* **81**, 2894-2897 (October 1998).

(8) A suggestion for nonlinear optics at near the single-photon level was described in: S. E. Harris and Y. Yamamoto, "Photon Switching by Quantum Interference," *Phys. Rev. Lett.* **81**, 3611-3614 (October 1998).

(9) The report of slow light was made in: L. V. Hau, S. E. Harris, Z. Dutton, and C. H. Behroozi, "Light Speed Reduction to 17 Meters Per Second in an Ultracold Atomic Gas," *Nature* **397**, 594-598 (February 1999).

(10) The use of EIT to efficiently generate vacuum ultraviolet radiation was described in: A. J. Merriam, S. J. Sharpe, H. Xia, D. Manuszak, G. Y. Yin, and S. E. Harris, "Efficient Gas-Phase Generation of Coherent Vacuum Ultraviolet Radiation," *Opt. Lett.* **24**, 625-627 (May 1999).

(11) The idea that EIT could be obtained in a medium with hyperfine structure was experimentally shown in: H. Xia, A. J. Merriam, S. J. Sharpe, G. Y. Yin, and S. E. Harris, "Electromagnetically Induced Transparency with Spectator Momenta," *Phys. Rev. A* **59**, R3190-R3193 (May 1999).

(12) A description of nonlinear optics with slow light was given in: S. E. Harris and L. V. Hau, "Nonlinear Optics at Low Light Levels," *Phys. Rev. Lett.* **82**, 4611-4614 (June 1999).

(13) A general extension of EIT to greatly detuned dispersive media was given in: D. D. Yavuz, A. V. Sokolov, and S. E. Harris, "Eigenvectors of a Raman Medium," *Phys. Rev. Lett.* **84**, 75-78 (January 2000).

(14) We have experimentally demonstrated the use of molecular deuterium in two driving lasers to demonstrate collinearly generation of mutually-coherent, equidistant sidebands covering 50,000 cm^{-1} of spectral bandwidth: A. V. Sokolov, D. R. Walker, D. D. Yavuz, G. Y. Yin, and S. E. Harris, "Raman Generation by Phased and Antiphased Molecular States," *Phys. Rev. Lett.* **85**, 562-565 (July 2000).

(15) EIT has been used to produce efficient nonlinear frequency in an all-resonant double-Λ system. At power densities which are characteristic of cw lasers, we obtain 30% conversion efficiency from 233 nm to the vacuum ultraviolet at 186 nm. [A. J. Merriam, S. J. Sharpe, M. Shverdin, D. Manuszak, G. Y. Yin, and S. E. Harris, "Efficient Nonlinear Frequency Conversion in an All-Resonant Double-L System," *Phys. Rev. Lett.* **84**, 5308–5311 (June 2000).]

(16) A special feature of slowly-moving light is the associated spatial compression. If an optical pulse is slowed down in the ratio of c/V_g, it is also spatially compressed by this same factor. This compression, in turn, leads to extremely large longitudinal gradient forces. The forces, in turn, result in new types of optical scattering as well as ballistic atomic motion and atom surfing. [S. E. Harris, "Ondermotive Forces with Slow Light," *Phys. Rev. Lett.* **85**, 4032-4035 (November 2000).]

(17) In work which will soon be published we have shown that the sidebands which are obtained from our new type of Raman generator may be phased so as to produce amplitude- or frequency-modulated light. The modulation frequency is equal to the fundamental vibrational frequency of molecular deuterium (2994 cm^{-1} ≈ 90 THz). [A. V. Sokolov, D. D. Yavuz, D. R. Walker, G. Y. Yin, and S. E. Harris, "Light Modulation at Molecular Frequencies," *Phys. Rev. A* (to be published).]

We attach abstracts of all papers which were published during this report period as Appendix A. Appendix B includes copies of four of the most significant publications.

IV. RELATION TO OTHER CONTRACTS

We note that the work reported here was jointly supported by the U.S. Air Force Office of Scientific Research, the U.S. Office of Naval Research, and the U.S. Army Research Office OSD Multidisciplinary University Research Initiative Program (MURI).

V. LIST OF PUBLICATIONS AND TECHNICAL REPORTS

1. Publications

S. E. Harris and A. V. Sokolov, "Subfemtosecond Pulse Generation by Molecular Modulation," *Phys. Rev. Lett.* **81**, 2894-2897 (October 1998).

S. E. Harris and Y. Yamamoto, "Photon Switching by Quantum Interference," *Phys. Rev. Lett.* **81**, 3611-3614 (October 1998).

L. V. Hau, S. E. Harris, Z. Dutton, and C. H. Behroozi, "Light Speed Reduction to 17 Meters Per Second in an Ultracold Atomic Gas," *Nature* **397**, 594-598 (February 1999).

A. V. Sokolov, D. D. Yavuz, and S. E. Harris, "Subfemtosecond Pulse Generation by Rotational Molecular Modulation," *Opt. Lett.* **24**, 557-559 (April 1999).

A. J. Merriam, S. J. Sharpe, H. Xia, D. Manuszak, G. Y. Yin, and S. E. Harris, "Efficient Gas-Phase Generation of Coherent Vacuum Ultraviolet Radiation," *Opt. Lett.* **24**, 625-627 (May 1999).

H. Xia, A. J. Merriam, S. J. Sharpe, G. Y. Yin, and S. E. Harris, "Electromagnetically Induced Transparency with Spectator Momenta," *Phys. Rev. A* **59**, R3190-R3193 (May 1999).

S. E. Harris and L. V. Hau, "Nonlinear Optics at Low Light Levels," *Phys. Rev. Lett.* **82**, 4611-4614 (June 1999).

F. L. Kien, J. Q. Liang, M. Katsuragawa, K. Ohtsuki, K. Hakuta, and A. V. Sokolov, "Subfemtosecond Pulse Generation with Molecular Coherence Control in Stimulated Raman Scattering," *Phys. Rev. A* **60**, 1562-1571 (August 1999).

A. V. Sokolov, "Subfemtosecond Compression of Periodic Laser Pulses," *Opt. Lett.* **24**, 1248-1250 (September 1999).

A. J. Merriam, S. J. Sharpe, H. Xia, D. Manuszak, G. Y. Yin, and S. E. Harris, "Efficient Gas-Phase VUV Frequency Up-Conversion," *IEEE J. Selected Topics in Quantum Electron.* **5**, 1502-1509 (November/December 1999).

D. D. Yavuz, A. V. Sokolov, and S. E. Harris, "Eigenvectors of a Raman Medium," *Phys. Rev. Lett.* **84**, 75-78 (January 2000).

A. V. Sokolov, D. R. Walker, D. D. Yavuz, G. Y. Yin, and S. E. Harris, "Raman Generation by Phased and Antiphased Molecular States," *Phys. Rev. Lett.* **85** 562–565 (July 2000).

A. J. Merriam, S. J. Sharpe, M. Shverdin, D. Manuszak, G. Y. Yin, and S. E. Harris, "Efficient Nonlinear Frequency Conversion in an All-Resonant Double-Λ System," *Phys. Rev. Lett.* **84**, 5308-5311 (June 2000).

S. E. Harris, "Pondermotive Forces with Slow Light" *Phys. Rev. Lett.* **85**, 4032–4035 (November 2000).

S. E. Harris, A. V. Sokolov, D. R. Walker, D. D. Yavuz, and G. Y. Yin, "Collinear Light Scattering Using Electromagnetically Induced Transparency, in *Atomic Physics 17 (ICAP 2000)*, E. Arimondo, D. Natale, and M. Inguscio, eds. (New York, American Institute of Physics, 2001), pp. 189-203.

A. V. Sokolov, D. D. Yavuz, D. R. Walker, G. Y. Yin, and S. E. Harris, "Light Modulation at Molecular Frequencies," Phys. Rev. A (to be published).

A. V. Sokolov, S. J. Sharpe, M. Shverdin, D. R. Walker, D. D. Yavuz, G. Y. Yin, and S. E. Harris, "Optical Frequency Conversion by a Rotating Molecular Waveplate," Opt. Lett. (to be published).

A. V. Sokolov, D. D. Yavuz, and S. E. Harris, "Refractive Index of a Strongly Driven Raman Medium" Phys. Rev. A (submitted for publication).

2. Technical Reports

S. E. Harris, "Research Studies on Electromagnetically Induced Transparency," Interim Progress Report (8 January 1998 - 31 December 1998).

S. E. Harris, "Research Studies on Electromagnetically Induced Transparency," Interim Progress Report (1 January 1999 - 31 December 1999).

L. V. Hau and S. E. Harris, "Slow Velocities and Nonlinear Optics in Cold Atoms" ARO Research Highlights Report (1999).

VI. LIST OF PARTICIPATING SCIENTIFIC PERSONNEL

Harris, S. E.	Principal Investigator
Yin, G. Y.	Senior Researcher
Epp, P. T.	Student (Ph.D. awarded March 2000)
Manuszak, D.	Student
Merriam, A. J.	Student (Ph.D. awarded June 2000)
Sharpe, S. J.	Student
Shverdin, M.	Student
Sokolov, A. V.	Student (Ph.D. submitted March 2001)
Yavuz, D.	Student

VII. REPORT ON INVENTIONS

"Method and Apparatus for Nonlinear Frequency Generation Using a Strongly-Driven Local Oscillator."

"Method and Apparatus for Single Longitudinal Mode Operation of Pulsed Laser Oscillators Using Efficient Self-Seeding."

VIII. BIBLIOGRAPHY OF PRINCIPAL INVESTIGATOR

1. S. E. Harris and A. E. Siegman, "A Proposed FM Phototube for Demodulating Microwave-Frequency-Modulated Light Signals," *IRE Trans. Electron Devices* **ED-9**, 322-329 (July 1962).

2. S. E. Harris, B. J. McMurtry, and A. E. Siegman, "Modulation and Direct Demodulation of Coherent and Incoherent Light at a Microwave Frequency," *Appl. Phys. Lett.* **1**, 37-39 (October 1962).

3. A. E. Siegman, S. E. Harris, and B. J. McMurtry, "Optical Heterodyning and Optical Demodulation at Microwave Frequencies," in *Optical Masers*, edited by Jerome Fox (New York, Polytechnic Institute of Brooklyn, 1963), pp. 511-527.

4. S. E. Harris, "Conversion of FM Light to AM Light Using Birefringent Crystals," *Appl. Phys. Lett.* **2**, 47-49 (January 1963).

5. A. E. Siegman, S. E. Harris, and B. J. McMurtry, "Microwave Demodulation of Light," in *Quantum Electronics III*, edited by P. Grivet and N. Bloembergen (New York, Columbia University Press, 1964), vol. 2, pp. 1651-1658.

6. S. E. Harris, "An FM-AM Optical Converter," in *Quantum Electronics III*, edited by P Grivet and N. Bloembergen (New York, Columbia University Press, 1964), vol. 2, pp. 1671-1677.

7. S. E. Harris, R. W. DeGrasse, and E. O. Schulz-DuBois, "Cutoff Frequencies of the Dielectrically Loaded Comb Structure as Used in Traveling-Wave Lasers," *Bell Syst. Tech. J.* **XLIII**, 437-484 (January 1964).

8. S. E. Harris and E. O. Ammann, "Optical Network Synthesis Using Birefringent Crystals," *Proc. IEEE* **52**, 411 (April 1964).

9. S. E. Harris, "Demodulation of Phase-Modulated Light Using Birefringent Crystals," *Proc. IEEE* **52**, 823-831 (July 1964).

10. S. E. Harris and A. E. Siegman, "A Technique for Optical Frequency Translation Utilizing the Quadratic Electrooptic Effect in Cubic Crystals," *Appl. Opt.* **3**, 1089-1090 (September 1964).

11. S. E. Harris, "Conversion of Frequency-Modulated Light to Space-Modulated Light," *J. Opt. Soc. Am.* **54**, 1147-1151 (September 1964).

12. S. E. Harris, E. O. Ammann, and I. C. Chang, "Optical Network Synthesis Using Birefringent Crystals. I. Synthesis of Lossless Networks of Equal-Length Crystals," *J. Opt. Soc. Am.* **54**, 1267-1279 (October 1964).

13. R. Targ, G. A. Massey, and S. E. Harris, "Laser Frequency Translation by Means of Electro-Optic Coupling Control," *Proc. IEEE* **52**, 1247 (October 1964).

14. S. E. Harris and Russell Targ, "FM Oscillation of the He-Ne Laser," *Appl. Phys. Lett.* **5**, 202-204 (November 1964).

15. S. E. Harris and O. P. McDuff, "FM Laser Oscillation - Theory," *Appl. Phys. Lett.* **5**, 205-206 (November 1964).

16. S. E. Harris, O. P. McDuff, and Russell Targ, "FM Laser Oscillation - Theory and Experiment," *IEEE International Convention Record* **5**, 21-26 (1965).

17. Stephen E. Harris, Gail A. Massey, M. Kenneth Oshman, and Russell Targ, "Controlling Laser Oscillation," *Electronics* **38**, 1-5 (September 1965).

18. S. E. Harris and O. P. McDuff, "Theory of FM Laser Oscillation," *IEEE J. Quantum Electron.* **QE-1**, 245-262 (September 1965).

19. S. E. Harris, M. Kenneth Oshman, B. J. McMurtry, and E. O. Ammann, "Proposed Frequency Stabilization of the FM Laser," *Appl. Phys. Lett.* **7**, 185-187 (October 1965).

20. S. E. Harris and B. J. McMurtry, "Frequency Selective Coupling to the FM Laser," *Appl. Phys. Lett.* **7**, 265-267 (November 1965).

21. S. E. Harris, "Proposed Backward Wave Oscillation in the Infrared," *Appl. Phys. Lett.* **9**, 114-116 (August 1966).

22. S. E. Harris, "Stabilization and Modulation of Laser Oscillators by Internal Time-Varying Perturbation," *Appl. Opt.* **5**, 1639-1651 (October 1966).

23. S. E. Harris, "Threshold of Multimode Parametric Oscillators," *IEEE J. Quantum Electron.* **QE-2**, 701 (October 1966).

24. O. P. McDuff and S. E. Harris, "Nonlinear Theory of the Internally Loss-Modulated Laser," *IEEE J. Quantum Electron.* **QE-3**, 101-111 (March 1967).

25. S. E. Harris, M. K. Oshman, and R. L. Byer, "Observation of Tunable Optical Parametric Fluorescence," *Phys. Rev. Lett.* **18**, 732-734 (May 1967).

26. S. E. Harris, "Threshold of Phase-Locked Parametric Oscillators," *IEEE J. Quantum Electron.* **QE-3**, 205 (May 1967).

27. R. L. Byer and S. E. Harris, "Power and Bandwidth of Spontaneous Parametric Emission," *Phys. Rev.* **168**, 1064-1068 (April 1968).

28. M. K. Oshman and S. E. Harris, "Theory of Optical Parametric Oscillation Internal to the Laser Cavity," *IEEE J. Quantum Electron.* **QE-4**, 491-502 (August 1968).

29. R. L. Byer, M. K. Oshman, J. F. Young, and S. E. Harris, "Visible CW Parametric Oscillator," *Appl. Phys. Lett.* **13**, 109-111 (August 1968).

30. C. M. McIntyre and S. E. Harris, "Achromatic Wave Plates for the Visible Spectrum," *J. Opt. Soc. Am.* **58**, 1575-1580 (December 1968).

31. R. L. Byer, S. E. Harris, D. J. Kuizenga, J. F. Young, and R. S. Feigelson, "Nonlinear Optical Properties of $Ba_2NaNb_5O_{15}$ in the Tetragonal Phase," *J. Appl. Phys.* **40**, 444 (January 1969).

32. S. E. Harris and R. W. Wallace, "Acousto-Optic Tunable Filter," *J. Opt. Soc. Am.* **59**, 744-747 (June 1969).

33. S. E. Harris, "Method to Lock an Optical Parametric Oscillator to an Atomic Transition," *Appl. Phys. Lett.* **14**, 335-337 (June 1969).

34. R. W. Wallace and S. E. Harris, "Oscillation and Doubling of the 0.946-μ Line in Nd^{3+}:YAG," *Appl. Phys. Lett.* **15**, 111-112 (August 1969).

35. S. E. Harris, S. T. K. Nieh, and D. K. Winslow, "Electronically Tunable Acousto-Optic Filter," *Appl. Phys. Lett.* **15**, 325-326 (November 1969).

36. S. E. Harris, "Tunable Optical Parametric Oscillators," *Proc. IEEE* **57**, 2096-2113 (December 1969).

37. James E. Murray and S. E. Harris, "Pulse Lengthening Via Overcoupled Internal Second-Harmonic Generation," *J. Appl. Phys.* **41**, 609-613 (February 1970).

38. S. E. Harris, S. T. K. Nieh, and R. S. Feigelson, "$CaMoO_4$ Electronically Tunable Optical Filter," *Appl. Phys. Lett.* **17**, 223-225 (September 1970).

39. Richard W. Wallace and Stephen E. Harris, "Extending the Tunability Spectrum," *Laser Focus* (November 1970), pp. 42-44.

40. J. F. Young, R. B. Miles, S. E. Harris, and R. W. Wallace, "Pump Linewidth Requirement for Optical Parametric Oscillators," *J. Appl. Phys.* **42**, 497-498 (January 1971).

41. J. F. Young, J. E. Murray, R. B. Miles, and S. E. Harris, "Q-Switched Laser With Controllable Pulse Length," *Appl. Phys. Lett.* **18**, 129-130 (February 1971).

42. D. J. Taylor, S. E. Harris, S. T. K. Nieh, and T. W. Hänsch, "Electronic Tuning of a Dye Laser Using the Acousto-Optic Filter," *Appl. Phys. Lett.* **19**, 269-271 (October 1971).

43. S. E. Harris and R. B. Miles, "Proposed Third-Harmonic Generation in Phase-Matched Metal Vapors," *Appl. Phys. Lett.* **19**, 385-387 (November 1971).

44. J. F. Young, G. C. Bjorklund, A. H. Kung, R. B. Miles, and S. E. Harris, "Third-Harmonic Generation in Phase-Matched Rb Vapor," *Phys. Rev. Lett.* **27**, 1551-1553 (December 1971).

45. S. T. K. Nieh and S. E. Harris, "Aperture-Bandwidth Characteristics of the Acousto-Optic Filter," *J. Opt. Soc. Am.* **62**, 672-676 (May 1972).

46. A. H. Kung, J. F. Young, G. C. Bjorklund, and S. E. Harris, "Generation of Vacuum Ultraviolet Radiation in Phase-Matched Cd Vapor," *Phys. Rev. Lett.* **29**, 985-988 (October 1972).

47. A. H. Kung, J. F. Young, and S. E. Harris, "Generation of 1182-Å Radiation in Phase-Matched Mixtures of Inert Gases," *Appl. Phys. Lett.* **22**, 301-302 (March 1973). [Erratum: *Appl. Phys. Lett.* **28**, 239 (February 1976).]

48. R. B. Miles and S. E. Harris, "Optical Third-Harmonic Generation in Alkali Metal Vapors," *IEEE J. Quantum Electron.* **QE-9**, 470-484 (April 1973).

49. S. E. Harris, "Generation of Vacuum-Ultraviolet and Soft-X-Ray Radiation Using High-Order Nonlinear Optical Polarizabilities," *Phys. Rev. Lett.* **31**, 341-344 (August 1973).

50. S. E. Harris, A. H. Kung, E. A. Stappaerts, and J. F. Young, "Stimulated Emission in Multiple-Photon-Pumped Xenon and Argon Excimers," *Appl. Phys. Lett.* **23**, 232-234 (September 1973).

51. S. E. Harris, J. F. Young, A. H. Kung, D. M. Bloom, and G. C. Bjorklund, "Generation of Ultraviolet and Vacuum Ultraviolet Radiation," in *Laser Spectroscopy*, edited by R. G. Brewer and A. Mooradian (New York, Plenum, 1974), pp. 59-75.

52. S. E. Harris and D. M. Bloom, "Resonantly Two-Photon Pumped Frequency Converter," *Appl. Phys. Lett.* **24**, 229-230 (March 1974).

53. D. M. Bloom, James T. Yardley, J. F. Young, and S. E. Harris, "Infrared Up-Conversion With Resonantly Two-Photon Pumped Metal Vapors," *Appl. Phys. Lett.* **24**, 427-428 (May 1974).

54. S. E. Harris and D. B. Lidow, "Nonlinear Optical Processes by van der Waals Interaction During Collision," *Phys. Rev. Lett.* **33**, 674-676 (September 1974). [Erratum: *Phys. Rev. Lett.* **34**, 172 (January 1975).]

55. G. C. Bjorklund, S. E. Harris, and J. F. Young, "Vacuum Ultraviolet Holography," *Appl. Phys. Lett.* **25**, 451-452 (October 1974).

56. S. E. Harris, J. F. Young, A. H. Kung, D. M. Bloom, and G. C. Bjorklund, "Generation of Ultraviolet and Vacuum Ultraviolet Radiation," in *Laser Applications to Optics and Spectroscopy*, edited by Stephen F. Jacobs, Murray Sargent III, James F. Scott, and Marlan O. Scully (Reading, MA, Addison-Wesley, 1975), pp. 181–197.

57. S. Haroche, J. C. Pebay-Peyroula, T. W. Hänsch, and S. E. Harris, Editors, *Laser Spectroscopy* (New York, Springer-Verlag, 1975).

58. S. E. Harris and D. B. Lidow, "Isotope Separation by Optically Pumped Ionizing Collisions," *Appl. Phys. Lett.* **26**, 104-105 (February 1975).

59. D. M. Bloom, G. W. Bekkers, J. F. Young, and S. E. Harris, "Third Harmonic Generation in Phase-Matched Alkali Metal Vapors," *Appl. Phys. Lett.* **26**, 687-689 (June 1975).

60. D. M. Bloom, J. F. Young, and S. E. Harris, "Mixed Metal Vapor Phase Matching for Third-Harmonic Generation," *Appl. Phys. Lett.* **27**, 390-392 (October 1975).

61. S. E. Harris, "Nonlinear Optical Techniques for Generation of VUV and Soft X-Ray Radiation," in *Very High Resolution Spectroscopy*, edited by R. A. Smith (New York, Academic Press, 1976), pp. 143-157.

62. S. E. Harris, R. W. Falcone, W. R. Green, D. B. Lidow, J. C. White, and J. F. Young, "Laser Induced Collisions," in *Tunable Lasers and Applications*, edited by A. Mooradian, T. Jaeger, and P. Stokseth (New York, Springer-Verlag, 1976), pp. 193-206.

63. K. S. Hsu, A. H. Kung, L. J. Zych, J. F. Young, and S. E. Harris, "1202.8-Å Generation in Hg Using a Parametrically Amplified Dye Laser," *IEEE J. Quantum Electron.* **QE-12**, 60-62 (January 1976).

64. D. B. Lidow, R. W. Falcone, J. F. Young, and S. E. Harris, "Inelastic Collision Induced by Intense Optical Radiation," *Phys. Rev. Lett.* **36**, 462-464 (March 1976). [Erratum: *Phys. Rev. Lett.* **37**, 1590 (December 1976).]

65. Eddy A. Stappaerts, Gerhard W. Bekkers, James F. Young, and Stephen E. Harris, "The Effect of Linewidth on the Efficiency of Two-Photon-Pumped Frequency Converters," *IEEE J. Quantum Electron.* **QE-12**, 330-333 (June 1976).

66. E. A. Stappaerts, S. E. Harris, and J. F. Young, "Efficient IR Image Up-Conversion in Two-Photon Resonantly Pumped Cs Vapor," *Appl. Phys. Lett.* **29**, 669-670 (November 1976).

67. J. F. Young and S. E. Harris, "Inelastic Collisions by Intense Laser Radiation," in *Multiphoton Processes*, edited by Joseph H. Eberly and Peter Lambropoulos (New York, Wiley, 1977), pp. 397-402.

68. R. W. Falcone, W. R. Green, J. C. White, J. F. Young, and S. E. Harris, "Observation of Laser-Induced Inelastic Collisions," *Phys. Rev. A* **15**, 1333-1335 (March 1977).

69. S. E. Harris, "Spontaneous Anti-Stokes Scattering as a High-Resolution and Picosecond-Time-Scale VUV Light Source," *Appl. Phys. Lett.* **31**, 498-500 (October 1977).

70. Stephen E. Harris and Jonathan C. White, "Numerical Analysis of Laser Induced Inelastic Collisions," *IEEE J. Quantum Electron.* **QE-13**, 972-978 (December 1977).

71. S. E. Harris, J. Lukasik, J. F. Young, and L. J. Zych, "Anti-Stokes Emission as a VUV and Soft X-Ray Source," in *Picosecond Phenomena*, edited by C. V. Shank, E. P. Ippen, and S. L. Shapiro (New York, Springer-Verlag, 1978), pp. 76-84.

72. Stephen E. Harris and James F. Young, "Rapid Laser Induced Energy Transfer in Atomic Systems," in *Radiation Energy Conversion in Space*, edited by Kenneth W. Billman (New York, American Institute of Aeronautics and Astronautics, 1978), pp. 569-582.

73. L. J. Zych, J. Lukasik, J. F. Young, and S. E. Harris, "Laser-Induced Two-Photon Blackbody Radiation in the Vacuum Ultraviolet," *Phys. Rev. Lett.* **40**, 1493-1496 (June 1978).

74. R. W. Falcone, J. R. Willison, J. F. Young, and S. E. Harris, "Measurement of the He $1s2s\ {}^1S_0$ Isotopic Shift Using a Tunable VUV Anti-Stokes Light Source," *Opt. Lett.* **3**, 162-163 (November 1978).

75. J. C. White, G. A. Zdasiuk, J. F. Young, and S. E. Harris, "Observation of Radiative Collisional Fluorescence," *Phys. Rev. Lett.* **41**, 1709-1712 (December 1978).

76. S. E. Harris and J. F. Young, "Techniques for Rapid Laser Induced Energy Transfer Using Metastable States," in *Proceedings of the International Conference on Lasers '78*, edited by Vincent J. Corcoran (McLean, VA, STS Press, 1979), pp. 679–688.

77. S. E. Harris, J. F. Young, W. R. Green, R. W. Falcone, J. Lukasik, J. C. White, J. R. Willison, M. D. Wright, and G. A. Zdasiuk, "Laser Induced Collisional and Radiative Energy Transfer," in *Laser Spectroscopy IV*, edited by H. Walther and K. W. Rothe (New York, Springer-Verlag, 1979), pp. 349-359.

78. W. R. Green, J. Lukasik, J. R. Willison, M. D. Wright, J. F. Young, and S. E. Harris, "Measurement of Large Cross Sections for Laser-Induced Collisions," *Phys. Rev. Lett.* **42**, 970-973 (April 1979).

79. J. C. White, G. A. Zdasiuk, J. F. Young, and S. E. Harris, "Observation of Atomic-Pair Absorption With an Incoherent Source," *Opt. Lett.* **4**, 137-139 (May 1979).

80. W. R. Green, M. D. Wright, J. F. Young, and S. E. Harris, "Laser-Induced Charge Transfer to an Excited Ionic State," *Phys. Rev. Lett.* **43**, 120-123 (July 1979).

81. W. R. Green, M. D. Wright, J. Lukasik, J. F. Young, and S. E. Harris, "Observation of a Laser-Induced Dipole-Quadrupole Collision," *Opt. Lett.* **4**, 265-267 (September 1979).

82. S. E. Harris, "Proposal for a 207-Å Laser in Lithium," *Opt. Lett.* **5**, 1-3 (January 1980).

83. J. R. Willison, R. W. Falcone, J. C. Wang, J. F. Young, and S. E. Harris, "Emission Spectra of Core Excited Even-Parity ^2P States of Neutral Lithium," *Phys. Rev. Lett.* **44**, 1125-1128 (April 1980).

84. S. E. Harris, J. F. Young, R. W. Falcone, W. R. Green, D. B. Lidow, J. Lukasik, J. C. White, M. D. Wright, and G. A. Zdasiuk, "Laser Induced Collisional Energy Transfer," in *Atomic Physics 7*, edited by Daniel Kleppner and Francis M. Pipkin (New York, Plenum, 1981), pp. 407-428.

85. S. E. Harris, R. W. Falcone, M. Gross, R. Normandin, K. D. Pedrotti, J. E. Rothenberg, J. C. Wang, J. R. Willison, and J. F. Young, "Anti-Stokes Scattering as an XUV Radiation Source," in *Laser Spectroscopy V*, edited by A. R. W. McKellar, T. Oka, and B. P. Stoicheff (New York, Springer-Verlag, 1981), pp. 437-445.

86. Joshua E. Rothenberg and Stephen E. Harris, "XUV Lasers by Quartet to Doublet Energy Transfer in Alkali Atoms," *IEEE J. Quantum Electron.* **QE-17**, 418-422 (March 1981).

87. A. J. Mendelsohn, R. Normandin, S. E. Harris, and J. F. Young, "A Microwave-Pumped XeCl* Laser," *Appl. Phys. Lett.* **38**, 603-605 (April 1981).

88. Joshua E. Rothenberg, J. F. Young, and S. E. Harris, "High-Resolution Extreme-Ultraviolet Spectroscopy of Potassium Using Anti-Stokes Radiation," *Opt. Lett.* **6**, 363-365 (August 1981).

89. M. D. Wright, D. M. O'Brien, J. F. Young, and S. E. Harris, "Laser-Induced Charge-Transfer Collisions of Calcium Ions With Strontium Atoms," *Phys. Rev. A* **24**, 1750-1755 (October 1981).

90. J. R. Willison, R. W. Falcone, J. F. Young, and S. E. Harris, "Laser Spectroscopy of Metastable Extreme-Ultraviolet Levels in Lithium Atoms and Ions," *Phys. Rev. Lett.* **47**, 1827-1829 (December 1981).

91. S. E. Harris, J. F. Young, R. W. Falcone, Joshua E. Rothenberg, J. R. Willison, and J. C. Wang, "Anti-Stokes Scattering as an XUV Radiation Source and Flashlamp," in *Laser Techniques for Extreme Ultraviolet Spectroscopy*, edited by T. J. McIlrath and R. R. Freeman (New York, AIP, 1982), pp. 137-152.

92. J. F. Young, S. E. Harris, P. J. K. Wisoff, and A. J. Mendelsohn, "Microwave Excitation of Excimer Lasers," *Laser Focus* (April 1982), pp. 63-67

93. S. E. Harris, R. W. Falcone, and D. M. O'Brien, "Proposal for High-Power Radiative-Collisional Lasers," *Opt. Lett.* **7**, 397-398 (September 1982).

94. S. E. Harris, J. F. Young, R. W. Falcone, J. E. Rothenberg, and J. R. Willison, "Laser Techniques for Spectroscopy of Core-Excited Atomic Levels," *J. Phys. (Paris)* **43**, 243-254 (November 1982).

95. P. J. K. Wisoff, A. J. Mendelsohn, S. E. Harris, and J. F. Young, "Improved Performance of the Microwave-Pumped XeCl Laser," *IEEE J. Quantum Electron.* **QE-18**, 1839-1840 (November 1982).

96. S. E. Harris, J. F. Young, R. G. Caro, R. W. Falcone, D. E. Holmgren, D. J. Walker, J. C. Wang, Joshua E. Rothenberg, and J. R. Willison, "Laser Techniques for Extreme Ultraviolet Spectroscopy," in *Laser Spectroscopy VI*, edited by H. P. Weber and W. Luthy (New York, Springer-Verlag, 1983), pp. 376-381.

97. R. G. Caro, J. C. Wang, R. W. Falcone, J. F. Young, and S. E. Harris, "Soft X-Ray Pumping of Metastable Levels of Li^+," *Appl. Phys. Lett.* **42**, 9-11 (January 1983).

98. J. C. Wang, R. G. Caro, and S. E. Harris, "Novel Short-Pulse Photoionization Electron Source: Li(1s2s2p)$^4P^0$ Deexcitation Measurements in a Plasma," *Phys. Rev. Lett.* **51**, 767-770 (August 1983).

99. Joshua E. Rothenberg, James F. Young, and Stephen E. Harris, "Spontaneous Raman Scattering as a High-Resolution XUV Radiation Source," *IEEE J. Quantum Electron.* **QE-19**, 1795-1804 (December 1983). [Correction: *IEEE J. Quantum Electron.* **QE-20**, 986 (August 1984).]

100. D. E. Holmgren, D. J. Walker, D. A. King, and S. E. Harris, "Grotrian Diagram of the Quartet System of Na I," in *Laser Techniques in the Extreme Ultraviolet*, edited by S. E. Harris and T. B. Lucatorto (New York, AIP, 1984), pp. 157-161.

101. R. G. Caro, J. C. Wang, J. F. Young, and S. E. Harris, "The Excitation of Metastable Extreme Ultraviolet Levels," in *Laser Techniques in the Extreme Ultraviolet*, edited by S. E. Harris and T. B. Lucatorto (New York, AIP, 1984), pp. 417-426.

102. D. E. Holmgren, D. J. Walker, and S. E. Harris, "Emission at 1091 Å in Neutral Core-Excited Cs," in *Laser Techniques in the Extreme Ultraviolet*, edited by S. E. Harris and T. B. Lucatorto (New York, AIP, 1984), pp. 496-501.

103. D. E. Holmgren, R. W. Falcone, D. J. Walker, and S. E. Harris, "Measurement of Lithium and Sodium Metastable Quartet Atoms in a Hollow-Cathode Discharge," *Opt. Lett.* **9**, 85-87 (March 1984).

104. S. E. Harris, D. J. Walker, R. G. Caro, A. J. Mendelson, and R. D. Cowan, "Quasi-Metastable Quartet Levels in Alkali-Like Atoms and Ions," *Opt. Lett.* **9**, 168-170 (May 1984).

105. R. G. Caro, J. C. Wang, J. F. Young, and S. E. Harris, "Use of Laser-Produced Soft X-Rays for the Production of Excited Metastable Ions," *Phys. Rev. A* **30**, 1407-1418 (September 1984).

106. S. E. Harris, R. G. Caro, R. W. Falcone, D. E. Holmgren, J. E. Rothenberg, D. J. Walker, J. C. Wang, J. R. Willison, and J. F. Young, "Metastability in the XUV: Lasers and Spectroscopy," in *Atomic Physics 9*, edited by Robert S. Van Dyck Jr. and E. Norval Fortson (Singapore, World Scientific Publishing Co., 1985), pp. 462-479.

107. S. E. Harris, J. F. Young, A. J. Mendelsohn, D. E. Holmgren, K. D. Pedrotti, and D. P. Dimiduk, "Quasi-Metastable Energy Levels and Applications," in *Laser Spectroscopy VII*, edited by T. W. Hänsch and Y. R. Shen (New York, Springer-Verlag, 1985), pp. 162-165.

108. D. E. Holmgren, D. J. Walker, D. A. King, and S. E. Harris, "Laser Spectroscopy of Na I Quartets," *Phys. Rev. A* **31**, 677-683 (February 1985).

109. R. G. Caro, J. C. Wang, J. F. Young, and S. E. Harris, "X-Ray Excitation of Energetic Metastable Levels in Atoms and Ions," *Opt. News* **11**, 9-11 (March 1985).

110. A. J. Mendelsohn and S. E. Harris, "Proposal for an Extreme-Ultraviolet Selective Autoionization Laser in Zn III," *Opt. Lett.* **10**, 128-130 (March 1985).

111. K. D. Pedrotti, A. J. Mendelsohn, R. W. Falcone, J. F. Young, and S. E. Harris, "Extreme-Ultraviolet Emission Spectra of Core-Excited Levels in Sodium and Magnesium," *J. Opt. Soc. Am. B* **2**, 1942-1947 (December 1985).

112. J. F. Young, J. J. Macklin, and S. E. Harris, "Ellipsoidal Focusing of Soft X-Rays for Longitudinally Pumping Short Wavelength Lasers," in *Short Wavelength Coherent Radiation: Generation and Applications*, edited by D. T. Attwood and J. Bokor (New York, AIP, 1986), pp. 86-88.

113. R. G. Caro, P. J. K. Wisoff, G. Y. Yin, D. J. Walker, M. H. Sher, C. P. J. Barty, J. F. Young, and S. E. Harris, "Soft X-Ray Pumping of Inner-Shell Excited Levels for Extreme Ultraviolet Lasers," in *Short Wavelength Coherent Radiation: Generation and Applications*, edited by D. T. Attwood and J. Bokor (New York, AIP, 1986), pp. 145-156.

114. D. P. Dimiduk, K. D. Pedrotti, J. F. Young, and S. E. Harris, "Laser Spectroscopy of the 109.1 nm Transition in Neutral Cs," in *Short Wavelength Coherent Radiation: Generation and Applications*, edited by D. T. Attwood and J. Bokor (New York, AIP, 1986), pp. 213-218.

115. S. E. Harris and R. G. Caro, "Shake-Up as a Mechanism for Vacuum-Ultraviolet Lasers," *Opt. Lett.* **11**, 10-11 (January 1986).

116. K. D. Pedrotti, D. P. Dimiduk, J. F. Young, and S. E. Harris, "Identification and Oscillator-Strength Measurement of the 109.1-nm Transition in Neutral Cs," *Opt. Lett.* **11**, 425-427 (July 1986).

117. D. J. Walker, R. G. Caro, and S. E. Harris, "Proposal for an Extreme-Ultraviolet Auger Laser at 63.8 nm in Cs III," *J. Opt. Soc. Am. B* **3**, 1515-1518 (November 1986).

118. S. E. Harris and J. K. Spong, "Laser Depletion Spectroscopy of Core-Excited Levels," in *Laser Spectroscopy VIII*, edited by W. Persson and S. Svanberg (New York, Springer-Verlag, 1987), pp. 458-461.

119. J. F. Young, J. J. Macklin, and S. E. Harris, "Grazing-Incidence Ellipsoidal Reflector for Longitudinally Pumping Short-Wavelength Lasers," *Opt. Lett.* **12**, 90–92 (February 1987).

120. A. J. Mendelsohn, C. P. J. Barty, M. H. Sher, J. F. Young, and S. E. Harris, "Emission Spectra of Quasimetastable Levels of Alkali-Metal Atoms," *Phys. Rev. A* **35**, 2095-2101 (March 1987).

121. S. E. Harris and J. F. Young, "Core-Excited Metastable Levels: Application to Spectroscopy, to the Generation of Picosecond Extreme-Ultraviolet Pulses, and to Lasers," *J. Opt. Soc. Am. B* **4**, 547-562 (April 1987).

122. Guang-Yu Yin, C. P. J. Barty, D. A. King, D. J. Walker, S. E. Harris, and J. F. Young, "Low-Energy Pumping of a 108.9-nm Xenon Auger Laser," *Opt. Lett.* **12**, 331-333 (May 1987).

123. J. K. Spong, J. D. Kmetec, S. C. Wallace, J. F. Young, and S. E. Harris, "Laser Spectroscopy of Core-Excited Levels of Neutral Rubidium," *Phys. Rev. Lett.* **58**, 2631-2634 (June 1987).

124. M. H. Sher, J. J. Macklin, J. F. Young, and S. E. Harris, "Saturation of the Xe III 109-nm Laser Using Traveling-Wave Laser-Produced-Plasma Excitation," *Opt. Lett.* **12**, 891-893 (November 1987).

125. D. J. Walker, C. P. J. Barty, G. Y. Yin, J. F. Young, and S. E. Harris, "Observation of Super Coster-Kronig-Pumped Gain in Zn III," *Opt. Lett.* **12**, 894-896 (November 1987).

126. C. P. J. Barty, D. A. King, G. Y. Yin, K. H. Hahn, J. E. Field, J. F. Young, and S. E. Harris, "12.8 eV Laser in Neutral Cesium," in *Short Wavelength Coherent Radiation: Generation and Applications*, edited by Roger W. Falcone and Janos Kirz (Washington DC, Optical Society of America, 1988), vol. 2, pp. 13-20.

127. J. D. Kmetec and S. E. Harris, "Targets for Efficient Femtosecond-Time-Scale X-Ray Generation," in *Short Wavelength Coherent Radiation: Generation and Applications*, edited by Roger W. Falcone and Janos Kirz (Washington DC, Optical Society of America, 1988), vol. 2, pp. 209-212.

128. S. E. Harris, "Non-Reciprocity of Autoionizing Interferences: Lasers Without Inversion," in *Short Wavelength Coherent Radiation: Generation and Applications*, edited by Roger W. Falcone and Janos Kirz (Washington DC, Optical Society of America, 1988), vol. 2, pp. 414-417.

129. S. E. Harris and J. D. Kmetec, "Mixed-Species Targets for Femtosecond-Time-Scale X-Ray Generation," *Phys. Rev. Lett.* **61**, 62-65 (July 1988).

130. C. P. J. Barty, D. A. King, G. Y. Yin, K. H. Hahn, J. E. Field, J. F. Young, and S. E. Harris, "12.8-eV Laser in Neutral Cesium," *Phys. Rev. Lett.* **61**, 2201-2204 (November 1988).

131. J. K. Spong, A. Imamoglu, R. Buffa, and S. E. Harris, "Laser-Depletion Spectroscopy of Core-Excited Levels of Neutral Rubidium," *Phys. Rev. A* **38**, 5617–5627 (December 1988).

132. S. E. Harris, A. Imamoglu, and J. J. Macklin, "Nonreciprocal Emissive and Absorptive Processes," in *Laser Spectroscopy IX*, edited by Michael S. Feld, John E. Thomas, and Aram Mooradian (San Diego, Academic Press, 1989), pp. 232-235.

133. J. K. Spong, A. Imamoglu, J. D. Kmetec, and S. E. Harris, "Extreme-Ultraviolet Fluorescence from Core-Excited Levels of Neutral Rubidium," *Opt. Lett.* **14**, 168-170 (February 1989).

134. S. E. Harris, "Lasers Without Inversion: Interference of Lifetime-Broadened Resonances," *Phys. Rev. Lett.* **62**, 1033-1036 (February 1989).

135. S. E. Harris and J. J. Macklin, "Lasers Without Inversion: Single-Atom Transient Response," *Phys. Rev. A* **40**, 4135-4137 (October 1989).

136. A. Imamoglu and S. E. Harris, "Lasers Without Inversion: Interference of Dressed Lifetime-Broadened States," *Opt. Lett.* **14**, 1344-1346 (December 1989).

137. A. Imamoglu, J. J. Macklin, and S. E. Harris, "Interference of Lifetime Broadened Resonances: Nonreciprocal Gain and Loss Profiles," in *New Frontiers in Quantum Electrodynamics and Quantum Optics*, edited by A. O. Barut (New York, Plenum, 1990), pp. 223-229.

138. C. P. J. Barty, G. Y. Yin, J. E. Field, S. J. Benerofe, J. F. Young, and S. E. Harris, "Photo-Electron and Photoionization Pumping of XUV Lasers by Laser Produced Plasmas," in *X-Ray Lasers 1990*, edited by G. J. Tallents (New York, Institute of Physics, 1990), pp. 21-28.

139. S. E. Harris, J. E. Field, and A. Imamoglu, "Nonlinear Optical Processes Using Electromagnetically Induced Transparency," *Phys. Rev. Lett.* **64**, 1107-1110 (March 1990).

140. K. H. Hahn, D. A. King, and S. E. Harris, "Nonlinear Generation of 104.8-nm Radiation Within an Absorption Window in Zinc," *Phys. Rev. Lett.* **65**, 2777-2779 (November 1990).

141. M. H. Sher, S. J. Benerofe, J. F. Young, and S. E. Harris, "A 2-Hz 109-nm Mirrorless Laser," *J. Opt. Soc. Am. B* **8**, 114-116 (January 1991).

142. A. Imamoglu, J. E. Field, and S. E. Harris, "Lasers Without Inversion: A Closed Lifetime Broadened System," *Phys. Rev. Lett.* **66**, 1154-1156 (March 1991).

143. K.-J. Boller, A. Imamoglu, and S. E. Harris, "Observation of Electromagnetically Induced Transparency," *Phys. Rev. Lett.* **66**, 2593-2596 (May 1991).

144. S. J. Benerofe, Guang-Yu Yin, C. P. J. Barty, J. F. Young, and S. E. Harris, "116-nm H_2 Laser Pumped by a Traveling-Wave Photoionization Electron Source," *Phys. Rev. Lett.* **66**, 3136-3139 (June 1991).

145. J. E. Field, K. H. Hahn, and S. E. Harris, "Observation of Electromagnetically Induced Transparency in Collisionally Broadened Lead Vapor," *Phys. Rev. Lett.* **67**, 3062-3065 (November 1991).

146. K.-J. Boller, A. Imamoglu, and S. E. Harris, "Electromagnetically Induced Transparency in Sr Vapor," in *Laser Spectroscopy*, edited by M. Ducloy, E. Giacobino, and G. Camy (New Jersey, World Scientific, 1992), pp. 295-300.

147. J. D. Kmetec, C. L. Gordon III, J. J. Macklin, B. E. Lemoff, G. S. Brown, and S. E. Harris, "MeV X-Ray Generation With a Femtosecond Laser," *Phys. Rev. Lett.* **68**, 1527-1530 (March 1992).

148. S. E. Harris, J. E. Field, and A. Kasapi, "Dispersive Properties of Electromagnetically Induced Transparency," *Phys. Rev. A* **46**, R29-R32 (July 1992).

149. C. P. J. Barty, G. Y. Yin, J. E. Field, D. A. King, K. H. Hahn, J. F. Young, and S. E. Harris, "Studies of a 96.9-nm Laser in Neutral Cesium," *Phys. Rev. A* **46**, 4286-4296 (October 1992).

150. S. J. Benerofe, G. Y. Yin, and S. E. Harris, "116 nm H_2 Laser Pumped by a Traveling-Wave Photoionization Electron Source," in *Vacuum Ultraviolet Radiation Physics*, edited by F. J. Wuilleumier, Y. Petroff, and I. Nenner (New Jersey, World Scientific, 1993), pp. 85-95.

151. S. E. Harris, "Electromagnetically Induced Transparency with Matched Pulses," *Phys. Rev. Lett.* **70**, 552-555 (February 1993).

152. Maneesh Jain, G. Y. Yin, J. E. Field, and S. E. Harris, "Observation of Electromagnetically Induced Phasematching," *Opt. Lett.* **18**, 998-1000 (June 1993).

153. S. E. Harris, J. J. Macklin, and T. W. Hänsch, "Atomic Scale Temporal Structure Inherent to High-Order Harmonic Generation," *Opt. Commun.* **100**, 487-490 (July 1993).

154. S. E. Harris, "Normal Modes for Electromagnetically Induced Transparency," *Phys. Rev. Lett.* **72**, 52-55 (January 1994).

155. C. P. J. Barty, C. L. Gordon III, J. D. Kmetec, B. E. Lemoff, and S. E. Harris, "Ultrashort High Peak Power Lasers and Generation of Hard Incoherent X-Rays," in *Proceedings of the International Conference on Lasers '92*, edited by C. P. Wang (McLean, VA, STS Press, 1993), pp. 44-51.

156. B. E. Lemoff, C. P. J. Barty, and S. E. Harris, "Femtosecond-Pulse-Driven, Electron-Excited XUV Lasers in Eight-Times-Ionized Noble Gases," *Opt. Lett.*. **19**, 569-671 (April 1994).

157. C. P. J. Barty, C. L. Gordon III, B. E. Lemoff, P. T. Epp, and S. E. Harris, "Ultrashort Pulse Terawatt Lasers for the Generation of Coherent and Incoherent X–Ray Sources," in *Proceedings of the International Conference on Lasers '93*, V. J. Corcoran and T. A. Goldman, eds. (McLean, VA, STS Press, 1994), pp. 196-202.

158. C. L. Gordon III, C. P. J. Barty, and S. E. Harris, "Time Gated X-Ray Imaging Using an Ultrashort Pulse, Laser-Produced-Plasma X-Ray Source, in *Proceedings of Ultrafast Phenomena IX*, P. F. Barbara, W. H. Knox, G. A. Mourou, and A. H. Zewail, eds. (New York, Springer-Verlag, 1994), pp. 278-279.

159. S. E. Harris, "Refractive Index Control with Strong Fields," *Opt. Lett.* **19**, 2018-2020 (December 1994).

160. G. Y. Yin, B. E. Lemoff, C. L. Gordon, III, C. P. J. Barty, and S. E. Harris, "A 10-Hz XUV Laser in Xe IX Driven by a Femtosecond Laser Source," in *Proceedings of the International Workshop on X-Ray Lasers and Optics*, H. S. Peng, ed. (China, High Power Laser and Particle Beams Journal Agency, 1995), pp. 40-48.

161. G. Y. Yin, B. E. Lemoff, C. L. Gordon, III, C. P. J. Barty, and S. E. Harris, "41.8-nm Laser in Xe IX Driven by a Femtosecond Laser Source," in *Proceedings of the International Conference on Lasers '94* , edited by V. J. Corcoran and T. A. Goldman (McLean, VA, STS Press, 1995), pp. 49-56.

162. B. E. Lemoff, G. Y. Yin, C. L. Gordon III, C. P. J. Barty, and S. E. Harris, "Demonstration of a 10-Hz, Femtosecond-Pulse-Driven XUV Laser at 41.8 nm in Xe IX," *Phys. Rev. Lett.* **74**, 1574-1577 (February 1995).

163. A. Kasapi, M. Jain, G. Y. Yin, and S. E. Harris, "Electromagnetically Induced Transparency: Propagation Dynamics," *Phys. Rev. Lett.* **74**, 2447-2450 (March 1995).

164. S. E. Harris and Z.-F. Luo, "Preparation Energy for Electromagnetically Induced Transparency," *Phys. Rev. A* **52** R928-R931 (August 1995).

165. S. M. Hooker and S. E. Harris, "Femtosecond-Pulse-Driven, Electron-Excited XUV Lasers in Be-Like Ions," *Opt. Lett.* **20**, 1994-1996 (October 1995).

166. M. Jain, A. J. Merriam, A. Kasapi, G. Y. Yin, and S. E. Harris, "Elimination of Optical Self-Focusing by Population Trapping," *Phys. Rev. Lett.* **75**, 4385-4388 (December 1995).

167. S. E. Harris, G. Y. Yin, A. Kasapi, M. Jain, and Z. F. Luo, "Electromagnetically Induced Transparency," in *Coherence and Quantum Optics VII*, J. Eberly, L. Mandel, and E. Wolf, eds. (New York, Plenum Press, 1996), pp. 295-304.

168. B. E. Lemoff, G. Y. Yin, C. L. Gordon, III, C. P. J. Barty, and S. E. Harris, "Femtosecond-Pulse-Driven, 10-Hz, 41.8-nm Laser in Xe IX," *J. Opt. Soc. Am. B* **13**, 180-184 (January 1996).

169. M. A. Kasevich and S. E. Harris, "Nonlinear Optical Properties of a Noninteracting Bose Gas," *Opt. Lett.* **21**, 677-679 (May 1996).

170. A. Kasapi, G. Y. Yin, M. Jain, and S. E. Harris, "Measurement of Lorentzian Linewidths by Pulse Propagation Delay," *Phys. Rev. A* **53**, 4547-4555 (June 1996).

171. M. Jain, H. Xia, G. Y. Yin, A. J. Merriam, and S. E. Harris, "Efficient Nonlinear Frequency Conversion with Maximal Atomic Coherence," *Phys. Rev. Lett.* **77**, 4326–4329 (November 1996).

172. S. E. Harris, "Electromagnetically Induced Transparency in an Ideal Plasma," *Phys. Rev. Lett.* **77**, 5357-5360 (December 1996).

173. S. E. Harris and M. Jain, "Optical Parametric Oscillators Pumped by Population Trapped Atoms," *Opt. Lett.* **22**, 636-638 (May 1997).

174. S. E. Harris and A. V. Sokolov, "Broadband Spectral Comb Generation with Refractive Index Control," *Phys. Rev.* A **55**, R4019-R4022 (June 1997).

175. S. E. Harris, G. Y. Yin, M. Jain, H. Xia, and A. J. Merriam, "Nonlinear Optics at Maximum Coherence," *Philos. Trans. R. Soc. London A* **355**, 2291-2304 (1997).

176. S. E. Harris, "Electromagnetically Induced Transparency," *Physics Today* **50**(7), 36-42 (July 1997).

177. P. T. Epp, S. M. Hooker, S. J. Sharpe, G. Y. Yin, and S. E. Harris, "Femtosecond-Pulse-Driven Short-Wavelength Lasers," *Proceedings of Lasers '96*, edited by V. J. Corcoran and T. A. Goldman (McLean, VA, STS Press, 1997), pp. 18-25.

178. G. Y. Yin, M. Jain, H. Xia, A. J. Merriam, and S. E. Harris, "Nonlinear Optical Frequency Conversion Using Electromagnetically Induced Transparency," in *Laser Spectroscopy XIII*, Z. J. Wang, Z. M. Zhang, and Y. Z. Wang, eds. (Singapore, World Scientific Publishing Co., 1998), pp. 343-348.

179. H. Xia, S. J. Sharpe, A. J. Merriam, and S. E. Harris, "Electromagnetically Induced Transparency in Atoms with Hyperfine Structure," *Phys. Rev. A* **56**, R3362-R3365 (November 1997).

180. A. V. Sokolov, G. Y. Yin, and S. E. Harris, "Nonlinear Optics at Maximum Coherence," in *Physics of Vibrations*, edited by F. V. Bunkin (Allerton Press, Inc., 1998), vol. 6, pp. 18-26.

181. S. E. Harris and A. V. Sokolov, "Subfemtosecond Pulse Generation by Molecular Modulation," *Phys. Rev. Lett.* **81**, 2894-2897 (October 1998).

182. S. E. Harris and Y. Yamamoto, "Photon Switching by Quantum Interference," *Phys. Rev. Lett.* **81**, 3611-3614 (October 1998).

183. L. V. Hau, S. E. Harris, Z. Dutton, and C. H. Behroozi, "Light Speed Reduction to 17 Meters Per Second in an Ultracold Atomic Gas," *Nature* **397**, 594-598 (February 1999).

184. A. V. Sokolov, D. D. Yavuz, and S. E. Harris, "Subfemtosecond Pulse Generation by Rotational Molecular Modulation," *Opt. Lett.* **24**, 557-559 (April 1999).

185. A. J. Merriam, S. J. Sharpe, H. Xia, D. Manuszak, G. Y. Yin, and S. E. Harris, "Efficient Gas-Phase Generation of Coherent Vacuum Ultraviolet Radiation," *Opt. Lett.* **24**, 625-627 (May 1999).

186. H. Xia, A. J. Merriam, S. J. Sharpe, G. Y. Yin, and S. E. Harris, "Electromagnetically Induced Transparency with Spectator Momenta," *Phys. Rev. A* **59**, R3190-R3193 (May 1999).

187. S. E. Harris and L. V. Hau, "Nonlinear Optics at Low Light Levels," *Phys. Rev. Lett.* **82**, 4611-4614 (June 1999).

188. A. J. Merriam, S. J. Sharpe, H. Xia, D. Manuszak, G. Y. Yin, and S. E. Harris, "Efficient Gas-Phase VUV Frequency Up-Conversion," *IEEE J. Selected Topics in Quantum Electron.* **5**, 1502-1509 (November/December 1999).

189. D. D. Yavuz, A. V. Sokolov, and S. E. Harris, "Eigenvectors of a Raman Medium," *Phys. Rev. Lett.* **84**, 75-78 (January 2000).

190. A. V. Sokolov, D. R. Walker, D. D. Yavuz, G. Y. Yin, and S. E. Harris, "Raman Generation by Phased and Antiphased Molecular States," *Phys. Rev. Lett.* **85** 562–565 (July 2000).

191. A. J. Merriam, S. J. Sharpe, M. Shverdin, D. Manuszak, G. Y. Yin, and S. E. Harris, "Efficient Nonlinear Frequency Conversion in an All-Resonant Double-Λ System," *Phys. Rev. Lett.* **84**, 5308-5311 (June 2000).

192. A. V. Sokolov, D. D. Yavuz, D. R. Walker, G. Y. Yin, and S. E. Harris, "Light Modulation at Molecular Frequencies," Phys. Rev. A (to be published).

193. S. E. Harris, "Pondermotive Forces with Slow Light" *Phys. Rev. Lett.* **85**, 4032–4035 (November 2000).

194. S. E. Harris, A. V. Sokolov, D. R. Walker, D. D. Yavuz, and G. Y. Yin, "Collinear Light Scattering Using Electromagnetically Induced Transparency," in *Atomic Physics 17*, edited by E. Arimondo, P. DeNatale, and M. Inguscio (American Institute of Physics, New York, 2001), pp. 189-203.

195. A. V. Sokolov, S. J. Sharpe, M. Shverdin, D. R. Walker, D. D. Yavuz, G. Y. Yin, and S. E. Harris, "Optical Frequency Conversion by a Rotating Molecular Waveplate," *Opt. Lett.* (to be published).

196. A. V. Sokolov, D. D. Yavuz, and S. E. Harris, "Refractive Index of a Strongly Driven Raman Medium" *Phys. Rev. A* (submitted for publication).

197. A. V. Sokolov, D. R. Walker, D. D. Yavuz, G. Y. Yin, and S. E. Harris, "Coherent Control of Multiphoton Ionization on a Few-Femtosecond Time Scale," *Phys. Rev. Lett.* (submitted for publication).

APPENDIX A

ABSTRACTS OF PUBLICATIONS

Subfemtosecond Pulse Generation by Molecular Modulation

S. E. Harris and A. V. Sokolov

Edward L. Ginzton Laboratory, Stanford University, Stanford, California 94305

(Received 18 May 1998)

We suggest a technique for producing subfemtosecond pulses of radiation. The technique is based on using electromagnetically induced transparency to produce a strongly driven molecular coherence. This coherence results in a Raman spectrum with Bessel function amplitudes and phases corresponding to a frequency modulated signal, thereby allowing compression by the group velocity dispersion of the same medium. [S0031-9007(98)07282-2]

PACS numbers: 42.65.Re, 32.80.Qk, 42.50.Gy, 42.65.Sf

Photon Switching by Quantum Interference

S. E. Harris and Y. Yamamoto

Edward L. Ginzton Laboratory, Stanford University, Stanford, California 94305

(Received 13 July 1998)

We describe a four-state atomic system that absorbs two photons, but does not absorb one photon. As a switch, in the ideal limit, it operates at an energy cost of one photon per switching event. [S0031-9007(98)07421-3]

PACS numbers: 32.80.–t, 42.50.Dv, 42.50.Gy, 42.50.Hz

Light speed reduction to 17 metres per second in an ultracold atomic gas

Lene Vestergaard Hau*†, S. E. Harris‡, Zachary Dutton*† & Cyrus H. Behroozi*§

* *Rowland Institute for Science, 100 Edwin H. Land Boulevard, Cambridge, Massachusetts 02142, USA*

† *Department of Physics, § Division of Engineering and Applied Sciences, Harvard University, Cambridge, Massachusetts 02138, USA*

‡ *Edward L. Ginzton Laboratory, Stanford University, Stanford, California 94305, USA*

Techniques that use quantum interference effects are being actively investigated to manipulate the optical properties of quantum systems[1]. One such example is electromagnetically induced transparency, a quantum effect that permits the propagation of light pulses through an otherwise opaque medium[2–5]. Here we report an experimental demonstration of electromagnetically induced transparency in an ultracold gas of sodium atoms, in which the optical pulses propagate at twenty million times slower than the speed of light in a vacuum. The gas is cooled to nanokelvin temperatures by laser and evaporative cooling[6–10]. The quantum interference controlling the optical properties of the medium is set up by a 'coupling' laser beam propagating at a right angle to the pulsed 'probe' beam. At nanokelvin temperatures, the variation of refractive index with probe frequency can be made very steep. In conjunction with the high atomic density, this results in the exceptionally low light speeds observed. By cooling the cloud below the transition temperature for Bose–Einstein condensation[11–13] (causing a macroscopic population of alkali atoms in the quantum ground state of the confining potential), we observe even lower pulse propagation velocities (17 m s^{-1}) owing to the increased atom density. We report an inferred nonlinear refractive index of 0.18 cm^2 W^{-1} and find that the system shows exceptionally large optical nonlinearities, which are of potential fundamental and technological interest for quantum optics.

Subfemtosecond pulse generation by rotational molecular modulation

A. V. Sokolov, D. D. Yavuz, and S. E. Harris

Edward L. Ginzton Laboratory, Stanford University, Stanford, California 94305

Received November 13, 1998

We extend a recent suggestion for the generation of subfemtosecond pulses by molecular modulation [Phys. Rev. Lett. **81**, 2894 (1998)] to the rotational spectrum of molecular hydrogen (H_2). When a rotational transition $|a\rangle \rightarrow |b\rangle$ is strongly driven ($|\rho_{ab}| = 0.5$) the generation and phase-slip lengths are of the same order and the Raman spectrum has approximately Bessel function sideband amplitudes. Numerical simulation predicts that this spectrum (generated in a 14-cm-long cell at 1-atm pressure of H_2) will compress into a train of pulses with 94-fs pulse separation and a pulse length of 0.5 fs.

OCIS codes: 320.0320, 320.5520, 190.3100, 030.1640.

Efficient gas-phase generation of coherent vacuum ultraviolet radiation

Andrew J. Merriam, S. J. Sharpe, H. Xia, D. Manuszak, G. Y. Yin, and S. E. Harris

Edward L. Ginzton Laboratory, Stanford University, Stanford, California 94305

Received February 1, 1999

We report the demonstration of a pulsed atomic lead (Pb) vapor-based vacuum ultraviolet frequency converter from 233 to 186 nm with unity photon-conversion efficiency. This conversion is attained without phase matching.

OCIS codes: 190.2620, 190.4410, 270.1670, 020.1670, 140.7220, 140.4380.

RAPID COMMUNICATIONS

PHYSICAL REVIEW A VOLUME 59, NUMBER 5 MAY 1999

Electromagnetically induced transparency with spectator momenta

Hui Xia, A. J. Merriam, S. J. Sharpe, G. Y. Yin, and S. E. Harris
Edward L. Ginzton Laboratory, Stanford University, Stanford, California 94305
(Received 18 May 1998)

We describe a method and present experimental results for obtaining electromagnetically induced transparency by adjusting the frequencies of two lasers so that they coincide with the centers of gravity of two hyperfine split transitions. No hyperfine states are in Raman resonance and the zero of the dipole moment results from the interference of the manifold of cooperating m states. [S1050-2947(99)50805-6]

PACS number(s): 42.50.Gy, 32.80.Qk, 33.15.Pw, 42.65.An,

Nonlinear Optics at Low Light Levels

S. E. Harris
Edward L. Ginzton Laboratory, Stanford University, Stanford, California 94305

Lene Vestergaard Hau
Rowland Institute for Science, 100 Edwin H. Land Boulevard, Cambridge, Massachusetts 02142
and Department of Physics, Harvard University, Cambridge, Massachusetts 02138
(Received 21 December 1998)

We show how the combination of electromagnetically induced transparency based nonlinear optics and cold atom technology, under conditions of ultraslow light propagation, allows nonlinear processes at energies of a few photons per atomic cross section. [S0031-9007(99)09290-X]

PACS numbers: 32.80.-t, 42.50.Dv, 42.50.Gy, 42.50.Hz

Subfemtosecond pulse generation with molecular coherence control in stimulated Raman scattering

Fam Le Kien,[1,2,*] J. Q. Liang,[1,2,3] M. Katsuragawa,[1,2,3] K. Ohtsuki,[3] and K. Hakuta[1,2,3]
[1]*Institute for Laser Science, University of Electro-Communications, Chofu, Tokyo 182-8585, Japan*
[2]*CREST, Japan Science and Technology Corporation, Kawaguchi, Saitama 332-0012, Japan*
[3]*Department of Applied Physics and Chemistry, University of Electro-Communications, Chofu, Tokyo 182-8585, Japan*

A. V. Sokolov
Edward L. Ginzton Laboratory, Stanford University, Stanford, California 94305
(Received 4 March 1999)

We study the generation of subfemtosecond pulses with the molecular coherence control in stimulated Raman scattering. We show analytically that the antiphased state temporally advances the higher frequencies with respect to the lower frequencies during a beating cycle. After some propagation distance, due to the dispersion and the difference between the antiphased and phased states in advancing high or low frequencies, the coherence is highest on the negative side of the detuning, when the two-photon Rabi frequency is about equal to the detuning. This asymmetry of the coherence magnitude with respect to the negative and positive sides of the Raman detuning is reflected in the behavior of the tuning characteristics of the high-order frequency components. When the Raman detuning is small, although the process is nonadiabatic, the subfemtosecond pulse generation may occur for both negative and positive sides of the detuning.
[S1050-2947(99)00908-7]

PACS number(s): 42.65.Re, 32.80.Qk, 42.50.Gy, 42.65.Sf

Subfemtosecond Compression of Periodic Laser Pulses

A. V. Sokolov

Edward L. Ginzton Laboratory, Stanford University, Stanford. California 94305

(April 23. 1999)

Abstract

We show that a recently proposed technique of molecular modulation can be used to compress mode-locked laser pulses into the subfemtosecond domain. The method relies on driving a molecular oscillation with a periodic pulse train. The coherent molecular motion. in turn. modulates the laser frequency and allows pulse compression by group velocity dispersion of the same molecular medium.

PACS numbers: 42.65.Re. 32.80.Qk, 42.50.Gy, 42.65.Sf

Efficient Gas-Phase VUV Frequency Up-Conversion

Andrew J. Merriam, Scott J. Sharpe, Hui Xia, Danielle A. Manuszak, G. Y. Yin, and Stephen E. Harris, *Fellow, IEEE*

***Abstract*—We describe a pulsed, atomic lead (Pb) vapor-based vacuum ultraviolet (VUV) frequency converter from 233 to 186 nm with near-unity photon conversion efficiency. This conversion is attained without phasematching and is accomplished by using electromagnetically induced transparency to drive a Raman transition to near-maximal coherence without loss or beam blow-up. Under these conditions, the linear and nonlinear polarizations of the generated 186-nm field are of the same order and complete conversion from the 233-nm field occurs within a single (nonphasematched) coherence length.**

***Index Terms*—Atomic physics, lead, nonlinearities, nonlinear optics, optical frequency conversion, optical parametric amplifiers, optical phasematching.**

Eigenvectors of a Raman Medium

D. D. Yavuz, A. V. Sokolov, and S. E. Harris
Edward L. Ginzton Laboratory, Stanford University, Stanford, California 94305
(Received 18 August 1999)

We show the existence of discrete sets of Raman sidebands which self-consistently establish a Raman coherence and propagate without change in amplitude and relative phase. Equivalently, there exist periodic femtosecond-time-scale, temporal pulse shapes which propagate without change in shape.

PACS numbers: 42.50.Gy, 32.80.Qk, 42.65.Dr, 42.65.Tg

Raman Generation by Phased and Antiphased Molecular States

A. V. Sokolov, D. R. Walker, D. D. Yavuz, G. Y. Yin, and S. E. Harris
Edward L. Ginzton Laboratory, Stanford University, Stanford, California 94305
(Received 14 December 1999)

We use molecular deuterium and two driving lasers to demonstrate collinear generation of mutually coherent equidistant sidebands, covering 50 000 cm^{-1} of spectral bandwidth and ranging from 2.94 μm to 195 nm in wavelength. The essential idea is the adiabatic preparation of a single, highly coherent ($|\rho_{ab}| = 0.33$) molecular eigenstate.

PACS numbers: 42.50.Gy, 32.80.Qk, 42.60.Fc, 42.65.Dr

Efficient Nonlinear Frequency Conversion in an All-Resonant Double-Λ System

Andrew J. Merriam, S. J. Sharpe, M. Shverdin, D. Manuszak, G. Y. Yin, and S. E. Harris
Edward L. Ginzton Laboratory, Stanford University, Stanford, California 94305
(Received 3 February 2000)

We demonstrate efficient, pulsed, gas-phase, nonlinear frequency conversion in a quadruply resonant, double-Λ system and, simultaneously, verify theoretical predictions of Rabi-frequency matching unique to absorbing nonlinear media. This system is used to up-convert ultraviolet light at 233 nm to the vacuum ultraviolet at 186 nm in atomic Pb vapor with small-signal conversion efficiencies exceeding 30% and with modest atomic density-length (NL) products (scale 10^{14} cm^{-2}) and optical power densities (10–100 kW/cm^2).

PACS numbers: 42.50.Gy, 42.50.Hz, 42.65.Ky, 42.79.Nv

Pondermotive Forces with Slow Light

S. E. Harris

Edward L. Ginzton Laboratory, Stanford University, Stanford. California 94305

(7/14/00)

Abstract

This work describes atomic processes which result from the greatly enhanced longitudinal gradient force which is inherent to the propagation of slow light. These processes are: (1) ballistic atom motion and atom surfing and (2) a type of local pondermotive nonlinearity or scattering which results from free-particle sinusoidal motion and the density variation caused by this motion.

PACS numbers: 32.80.-t, 32.80.Lg, 32.80.Qk, 42.50.Vk

Proceedings of
ICAP 2000

Collinear Light Scattering Using Electromagnetically Induced Transparency

S. E. Harris, A. V. Sokolov, D. R. Walker,
D. D. Yavuz, and G. Y. Yin

Edward L. Ginzton Laboratory,
Stanford University, Stanford, California 94305

Abstract. The paper describes two types of nonlinear optical processes which are based on electromagnetically induced transparency. These are: (1) Collinear generation of FM-like Raman sidebands and (2) a type of pondermotive light scattering which is inherent to the interaction of slow light with cold atoms. Connections to other areas of EIT-based nonlinear optics are also described.

Light Modulation at Molecular Frequencies

A. V. Sokolov, D. D. Yavuz, D. R. Walker, G. Y. Yin, and S. E. Harris

Edward L. Ginzton Laboratory, Stanford University, Stanford, California 94305

(June 2, 2000)

Abstract

We describe the generation of amplitude and frequency modulated light with a modulation frequency equal to the fundamental vibrational frequency of molecular deuterium (2994 $cm^{-1} \approx 90$ THz). This is a first step toward the synthesis of subfemtosecond pulses with prescribed temporal shape.

PACS numbers: 42.50.Gy, 32.80.Qk, 42.60.Fc, 42.65.Dr, 42.65.Re, 42.65.Sf

Optical Frequency Conversion by a Rotating Molecular Waveplate

A. V. Sokolov, S.J. Sharpe, M. Shverdin, D. R. Walker, D. D. Yavuz, G. Y. Yin, and S. E. Harris

Edward L. Ginzton Laboratory, Stanford University, Stanford, California 94305

(September 6, 2000)

Abstract

We demonstrate efficient laser frequency conversion in low-pressure molecular deuterium. We use two laser fields, with opposite circular polarizations, to produce a strong excitation of a ro-vibrational transition, at a frequency of 3167 cm^{-1}. The coherent molecular motion, in turn, modulates a third laser field (also circularly polarized) and results in highly efficient single-sideband conversion.

OCIS numbers: 190.2620, 190.5650, 270.1670, 300.6270

Refractive Index of a Strongly Driven Raman Medium

A. V. Sokolov, D. D. Yavuz, and S. E. Harris

Edward L. Ginzton Laboratory, Stanford University, Stanford, California 94305

(December 14, 2000)

Abstract

We derive an expression for the refractive index and self-focusing (or defocusing) distance in a strongly driven Raman medium, when the coherence of the molecular transition is on the order of its maximum value $|\rho_{ab}| \approx 0.5$. In this regime Raman generation and sideband propagation are intrinsically interrelated, and the refractive index is determined by the strong interaction of the whole comb of Raman sidebands with the medium.

PACS numbers: 42.65.Re, 32.80.Qk, 42.50.Gy, 42.65.Sf

APPENDIX B

SELECTED MOST IMPORTANT PUBLICATIONS

Subfemtosecond Pulse Generation by Molecular Modulation

S. E. Harris and A. V. Sokolov
Edward L. Ginzton Laboratory, Stanford University, Stanford, California 94305
(Received 18 May 1998)

We suggest a technique for producing subfemtosecond pulses of radiation. The technique is based on using electromagnetically induced transparency to produce a strongly driven molecular coherence. This coherence results in a Raman spectrum with Bessel function amplitudes and phases corresponding to a frequency modulated signal, thereby allowing compression by the group velocity dispersion of the same medium. [S0031-9007(98)07282-2]

PACS numbers: 42.65.Re, 32.80.Qk, 42.50.Gy, 42.65.Sf

This Letter suggests and analyzes a technique for producing subfemtosecond pulses of radiation. We will show that two laser beams whose frequency difference is slightly offset from a molecular transition will, for an appropriate choice of gas pressure and cell length, generate a spectrum of Raman sidebands whose Fourier transform is a periodic train of subfemtosecond pulses. The essence of the technique is the concurrent generation of a frequency modulated (FM) waveform and the use of group velocity dispersion to temporally compress this waveform. By numerically modeling this process in molecular deuterium (D_2), we calculate a generated train of pulses with a pulse spacing of 11.1 fs and a pulse length of 0.21 fs.

The coherence ρ_{ab} (Fig. 1) of the driven molecular transition is central to this work. This coherence is established by driving the molecular transition slightly off resonance with driving lasers of sufficient intensity that the product of their Rabi frequencies exceeds the product of the detuning from the molecular electronic states and the detuning from the Raman transition. With the linewidth of the applied laser pulses small as compared to the Raman detuning, the magnitude of the molecular coherence approaches 0.5, and its sign is determined by the sign of the Raman detuning.

For the conditions of the previous paragraph, the generation and phase-slip lengths are of the same order, and Raman sideband generation proceeds collinearly and very differently than in the conventional low coherence regime. In essence, the molecular motion now modulates the electronic refractive index much in the same way that a low frequency electric field modulates the refractive index of a polar crystal. Incident optical frequencies become frequency modulated with peak (sinusoidal) frequency deviations and spectral bandwidths that substantially exceed the width of the visible spectrum. This allows the extension of compression techniques, such as the use of group velocity dispersion as described here, to a new regime of short pulse compression.

In pertinent prior work, (1) the authors have noted how an electromagnetically induced transparency (EIT)-like excitation may be used to generate a broad comb of coherent sidebands, but the Bessel-function nature of the spectrum and the possibility of pulse compression were not observed [1]. (2) Hakuta and colleagues have experimentally demonstrated the generation of collinear Raman sidebands in solid molecular hydrogen [2]. (3) Kawano *et al.* have demonstrated the use of stimulated rotational Raman scattering to generate a broad spectrum and discussed possibilities for mode locking this spectrum [3]. (4) There has been considerable work in the area of on-axis Raman sideband generation [4]. (5) There is also some relation to the work of Kaplan and Shkolnikov on Raman solitons [5]. (6) Workers in the field of high-order harmonic generation have noted the possibility of obtaining a short time structure [6]. (But, in high-order harmonic generation, less than one part in 10^{-4} of the incident pulse energy is converted into the phased spectrum; by the approach of this work, near-complete conversion is likely.) (7) There is now considerable work on EIT [7] and related subjects [8]. (8) It has been shown that, by using a large atomic coherence, nearly complete frequency conversion will occur in a single coherence length [9].

The body of this Letter divides into four parts: (a) a summary of the formalism describing the interaction of the multilevel system of Fig. 1 with a comb of Raman

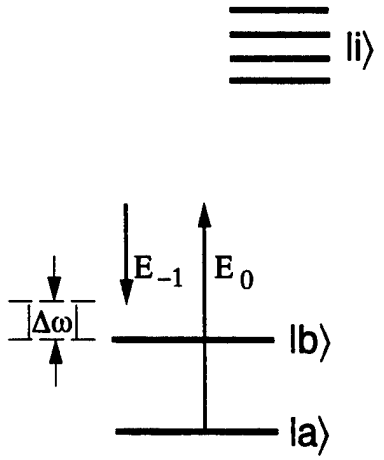


FIG. 1. Energy level schematic for establishing coherence ρ_{ab} in a molecular system. Laser fields are applied at the frequencies of the $q = 0$ and $q = -1$ sidebands. Here, the detuning $\Delta\omega$ from Raman resonance, as is required for establishing the antiphased state, is negative.

 0031-9007/98/81(14)/2894(4)$15.00

sidebands [1], this formalism includes all dispersive effects; (b) the FM solution for the ideal case of negligible dispersion; (c) a numerical simulation showing concurrent spectral generation and pulse compression in D_2; (d) a brief discussion of measurement possibilities.

We will work with the envelope quantities $E_q(z,t)$ and $\rho_{ab}(z,t)$ for electromagnetic fields and density matrix elements such that the full time and space quantities are $\hat{E}_q(z,t) = \mathrm{Re}\{E_q(z,t)\exp[j(\omega_q t - k_q z)]\}$ and $\hat{\rho}_{ab}(z,t) = \mathrm{Re}\{\rho_{ab}(z,t)\exp[j(\omega_m t - k_m z)]\}$, with $\omega_q = \omega_0 + q(\omega_b - \omega_a - \Delta\omega) = \omega_0 + q\omega_m$. The modulation frequency ω_m or, equivalently, the frequency difference between adjacent sidebands, is equal to the difference of the two applied frequencies ω_0 and ω_{-1}. The two-photon detuning $\Delta\omega$ is the difference of ω_m and the Raman transition frequency and, as shown in Fig. 1, is negative; $k_q = \omega_q/c$ and $k_m = \omega_m/c$.

We allow for an arbitrary number of virtual states $|\iota\rangle$ with energies $\hbar\omega_\iota$. The matrix elements from states $|a\rangle$ and $|b\rangle$ to these states are $\mu_{a\iota}$ and $\mu_{b\iota}$, respectively. When the derivatives of the probability amplitudes of the upper states $|\iota\rangle$ are small as compared to the detunings from these states, the problem can be written in terms of an effective, distance-dependent, two-by-two Hamiltonian [1]:

$$H_{\mathrm{eff}} = -\frac{1}{2}\begin{bmatrix} A & B\exp(-j\frac{\omega_m}{c}z) \\ C\exp(j\frac{\omega_m}{c}z) & D - 2\Delta\omega \end{bmatrix}, \quad (1)$$

where $A = \sum_q a_q|E_q|^2$; $B = \sum_q b_q E_q E_{q-1}^*$; $C = B^*$; and $D = \sum_q d_q|E_q|^2$. All rotating and nonrotating, as well as cross-transition, terms are retained within this Hamiltonian. We also assume the ideal case of zero linewidth for the $|a\rangle$-$|b\rangle$ transition. The constants a_q, b_q, and d_q determine the dispersion and coupling and are

$$\begin{aligned} a_q &= \frac{1}{2\hbar^2}\sum_i\left[\frac{|\mu_{ai}|^2}{(\omega_i - \omega_a) - \omega_q} + \frac{|\mu_{ai}|^2}{(\omega_i - \omega_a) + \omega_q}\right], \\ b_q &= \frac{1}{2\hbar^2}\sum_i\left[\frac{\mu_{ai}\mu_{bi}}{(\omega_i - \omega_a) - \omega_q} + \frac{\mu_{ai}\mu_{bi}}{(\omega_i - \omega_b) + \omega_q}\right], \\ d_q &= \frac{1}{2\hbar^2}\sum_i\left[\frac{|\mu_{bi}|^2}{(\omega_i - \omega_b) - \omega_q} + \frac{|\mu_{bi}|^2}{(\omega_i - \omega_b) + \omega_q}\right]. \end{aligned} \quad (2)$$

We assume that all of the molecular population is initially in the ground state and the envelopes of the exciting laser pulses vary slowly as compared to the separation of the eigenvalues of the Hamiltonian of Eq. (1). This allows adiabatic preparation of all of the molecules in one eigenstate. By choosing the sign of the detuning $\Delta\omega$, either the phased or antiphased eigenstate is selected at the input of the medium and is, as shown below, maintained throughout the medium. The molecular coherence ρ_{ab} for the phased state has the same sign as the quantity B in Eq. (1) and, for the antiphased state, has the opposite sign. Both eigenstates lead to an FM signal but, as shown below, in a normally dispersive medium, with the boundary condition of two incident frequencies, only the antiphased state allows pulse compression.

Defining

$$B = |B|\exp(j\varphi); \qquad \tan\theta = \frac{2|B|}{2\Delta\omega - D + A}, \quad (3)$$

the eigenstate, which adiabatically evolves from the ground state as field amplitudes are increased, and the coherence, which is obtained from it, are as follows:

$$\begin{aligned} |+\rangle &= \cos\frac{\theta}{2}\exp\left(j\frac{\varphi}{2}\right)|a\rangle \\ &\quad + \sin\frac{\theta}{2}\exp\left(-j\frac{\varphi}{2}\right)|b\rangle, \\ \rho_{ab} &= \frac{1}{2}\sin\theta\exp(j\varphi), \end{aligned} \quad (4)$$

where positive θ corresponds to the phased state and negative θ to the antiphased state. By choosing the intensities of the applied fields and the detuning $\Delta\omega$, the quantity $\sin\theta$ can be set to any value between -1 and 1.

The propagation equation for the qth sideband in local time is

$$\begin{aligned} \frac{\partial E_q}{\partial z} &= -j\eta\hbar\omega_q N(a_q\rho_{aa}E_q + d_q\rho_{bb}E_q + b_q\rho_{ab}E_{q-1} \\ &\quad + c_q\rho_{ab}^*E_{q+1}), \end{aligned} \quad (5)$$

where N is the number of molecules per volume and $\eta = (\mu/\epsilon_0)^{1/2}$.

In order to understand the numerical results which follow, we first exam the analytically solvable case of negligible dispersion and limited modulation bandwidth. We take all of the sidebands to be sufficiently far from the resonances that, in Eq. (2), $a_q = b_q = c_q = d_q \equiv a_0$. We define propagation constants $\beta_q = \eta\hbar\omega_q N a_0$ and change variables by $E_q = \tilde{E}_q\exp(-j\beta_q z)$, $\rho_{ab} = \tilde{\rho}_{ab}\exp(-j\beta_m z)$. With these approximations and definitions, Eq. (5) becomes

$$\frac{\partial\tilde{E}_q}{\partial z} = -j\beta_q(\tilde{\rho}_{ab}\tilde{E}_{q-1} + \tilde{\rho}_{ab}^*\tilde{E}_{q+1}). \quad (6)$$

The quantity $\tilde{\rho}_{ab} = \frac{1}{2}\sin\tilde{\theta}\exp(j\tilde{\varphi})$, where $\tilde{\theta}$ and $\tilde{\varphi}$ are given by Eq. (3) with E_q replaced by $\tilde{E}_q$. As the boundary conditions for the propagation equation, we assume that the fields $E_0(0)$ and $E_{-1}(0)$ are applied at the input of the medium and all other Raman sidebands are generated in the medium.

Although we will not make this approximation in the numerical work which follows, here we assume a restricted modulation bandwidth such that all ω_q are equal to ω_0 and $\beta_q = \beta_0$. By using the Bessel-function identities $2\frac{\partial J_n(x)}{\partial x} = [J_{n-1}(x) - J_{n+1}(x)]$ and $\sum_n J_{n+q}(x)J_{n+p}(x) = \delta_{pq}$, it may be verified that, with $\gamma = \beta_0\sin[\theta(0)]$, the solution $\tilde{\rho}_{ab}(z) = \rho_{ab}(0)$ and

$$\tilde{E}_q(z) = E_0(0)\exp\left[j\left(\varphi(0) - \frac{\pi}{2}\right)q\right]J_q(\gamma z) + E_{-1}(0)\exp\left[j\left(\varphi(0) - \frac{\pi}{2}\right)(q+1)\right]J_{q+1}(\gamma z) \quad (7)$$

satisfies both density matrix and propagation equations and also the boundary condition at $z = 0$. [$\varphi(0)$ is obtained from Eq. (3).] With $\sum_n J_n(x)\exp(jn\omega t) = \exp[jx\sin(\omega t)]$, the time domain signal is

$$E(t) = \mathrm{Re}\{E_0(0)\exp j[\omega_0 t + \gamma z\cos(\omega_m t + \varphi(0))] + E_{-1}(0)\exp j[\omega_{-1} t + \gamma z\cos(\omega_m t + \varphi(0))]\}. \quad (8)$$

Equation (8) is a superposition of two FM signals with center frequencies corresponding to the frequencies which are applied at $z = 0$ and a peak phase deviation γz. It is important that the frequency modulation leaves the eigenvectors of the Hamiltonian unchanged.

The total number of sidebands generated in a cell of length L is approximately equal to $2\gamma L$. The assumption of limited modulation bandwidth ($\omega_q = \omega_0$) implies that $\gamma z \ll \omega_0/\omega_m$; i.e., the peak frequency excursion must be small as compared to the center frequency. We have checked that, when this assumption is satisfied, the solution of Eq. (7) coincides with the exact numerical solution of Eq. (6).

Equations (7) and (8) hold for both the antiphased ($\sin\theta < 0$) and the phased case ($\sin\theta > 0$). But, in the antiphased case, the higher frequencies (blue) are temporally advanced with regard to the lower frequencies (red), while, in the phased case, red is advanced as compared to blue. When $E_0(0) \sim E_{-1}(0)$, only the signal generated by the antiphased state allows pulse compression in a normally dispersive medium. Figure 2(a) shows the time domain signal produced by the antiphased state; i.e., an FM signal with an envelope at the beat frequency ω_m.

Pulse compression is obtained by setting the group velocity delay which corresponds to the frequency deviation $\omega_d = \omega_m\gamma L$ equal to $\pi/2\omega_m$. Here, but not in the numerical work which follows, we assume a frequency invariant group velocity dispersion $\partial^2 k/\partial\omega^2$. The medium length l which is necessary for pulse compression is then determined by

$$\left.\frac{\partial^2 k}{\partial\omega^2}\right|_{\omega=\omega_0} l = \frac{\pi}{2\omega_m^2\gamma L}. \quad (9)$$

Figure 2(b) shows the signal which is obtained when the signal of Fig. 2(a) is transmitted through a medium of this length. Pulse compression with a ratio of pulse width to pulse separation of about $1/2\gamma L$ is observed.

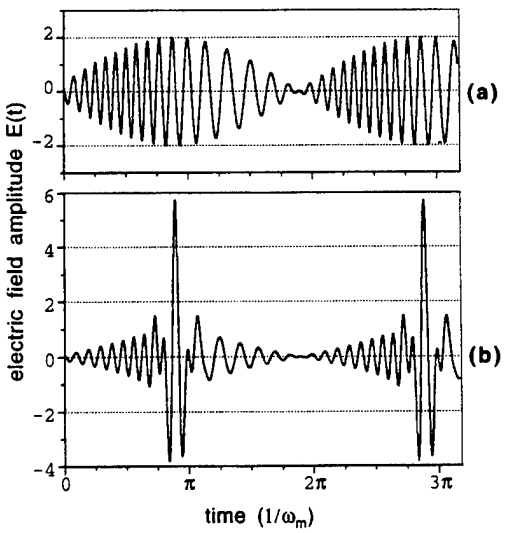


FIG. 2. Electric field amplitude vs time (a) as produced by an antiphased state without pulse compression. The parameters are $E_0(0) = E_{-1}(0) = 1$, $\omega_0/\omega_m = 18$, and $\gamma L = 7$. (b) Following pulse compression in a medium with a frequency-invariant group velocity dispersion and a length determined by Eq. (9).

We turn next to the numerical solution for D_2. We do not make any of the dispersive approximations described above. We find that both generation and pulse compression may be accomplished concurrently in the same medium. We assume E_0 and E_{-1} to be applied at $z = 0$ and retain sidebands between $q = -9$ and $q = 18$. The solution of Eq. (5) is obtained by forward stepping from $z = 0$ with the density matrix elements recalculated at each step.

We consider the fundamental vibrational transition in D_2 with $\omega_m = 2994\ \mathrm{cm}^{-1}$ and take all molecules in the $J = 0$ ground state. The constants a_q, b_q, and d_q include the contributions of the $0 - 52$nd vibrational transitions of the Lyman band and the $0 - 18$th transitions of the Werner band. Oscillator strengths and Franck-Condon factors are obtained from Allison and Dalgarno and energies are obtained from Herzberg [10]. The D_2 density and the total cell length are chosen such that the zeroth sideband, if propagating alone, would accumulate a phase shift $\beta_0 L = 14\pi$ rad. At a pressure of 1 atm, this corresponds to $L = 3.6$ cm.

We envision using single-mode pulsed lasers with pulse lengths of ~5 ns and pulse energies of ~50 mJ. We take the applied laser frequencies to be $\omega = 28\,183\ \mathrm{cm}^{-1}$ (frequency-tripled Nd:YAG) and $\omega_{-1} = 25\,189\ \mathrm{cm}^{-1}$ (frequency-doubled Ti:sapphire). We choose the Raman detuning $\Delta\omega = -1$ GHz to be much larger than both the Raman linewidth (~0.3 GHz for D_2 at STP) and the laser linewidth (~0.1 GHz for a single-mode, 5-ns-pulse laser). We set the power densities of E_0 and E_{-1} at $z = 0$ to $10^{11}\ \mathrm{W/cm}^2$. Evaluating Eqs. (1)–(4), $\sin\theta(0) \approx -0.9$. The coherence $|\rho_{ab}| \approx 0.45$ and the cell length are set such that about twenty sidebands are generated, and the concurrent group velocity delay is correct for pulse compression.

Figure 3 shows spectral and temporal evolution as a function of distance for the conditions of the previous

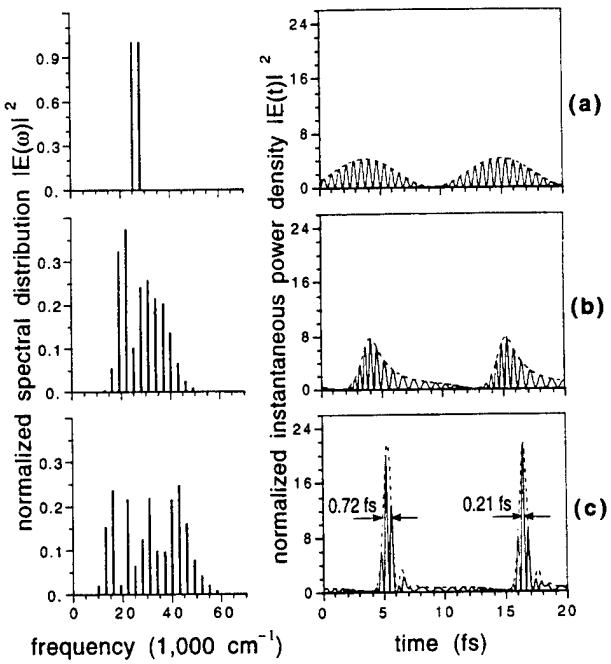


FIG. 3. Frequency spectrum (left) and instantaneous power density vs time (right) in D_2 at 1 atm pressure with applied power densities at $z = 0$ such that $|\rho_{ab}| \approx 0.45$. Parts (a)–(c) show the spectrum and temporal waveform for (a) $z = 0$ cm, (b) $z = 1.8$ cm, and (c) $z = 3.6$ cm. Sideband powers are normalized to those of the incident fields. The dashed lines on the time domain plots show the envelopes of possible single-cycle pulses.

paragraph. Sideband frequencies are predicted to range from about 1.4 μm in the infrared to 150 nm in the vacuum ultraviolet. At the appropriate cell length these frequencies synthesize a train of pulses with a pulse separation of 11.1 fs and a pulse length of 0.21 fs. We find sharp pulse compression at experimental conditions which are readily realizable. Because the ratio of the applied optical frequencies to the Raman transition frequency is, in general, noninteger, the peak of the optical cycle will drift from pulse to pulse within a narrow envelope. This envelope (width = 0.72 fs) is shown by the dashed line in each of the time domain plots of Fig. 3. For the numerical analysis we have treated the driving fields as monochromatic. For the pulsed excitation, as described above, we expect these results to apply for a time about equal to the dephasing time of the Raman transition, and therefore expect nearly all of the incident power to be converted into the periodic train of subfemtosecond pulses.

The predictions of this numerical analysis should be verifiable by two methods. The relative intensities of the sidebands could be measured; these differ in the phased and antiphased states, and are correlated with the phase of each sideband. In the time domain, the peak power density grows on a scale of 3 cm and varies in a predictable manner thereafter. In the region of the highest peak power (~5 mm long), one might expect multiphoton processes in either D_2 itself or in a dopant to light up as an indication of the presence of ultrashort pulses. The physics of such processes, as well as other nonlinear behavior in this regime is, in its own right, of considerable interest.

The principle approximations of the numerical results given here are the neglect of the dephasing of the Raman transition, the assumption that the driving fields are both quasimonochromatic and of an infinite extent in the transverse direction, and that all molecules are initially in the $J = 0$ ground state. Although the effects of self-focusing, temporal chirping, saturation of the Raman transition, and thermal population have not been discussed, we do not see contradictions.

In summary, we have described how the ideas of frequency modulation and pulse compression may be extended to the atomic time scale. The technique will allow the generation of pulses of high intensity radiation with a pulse length which is less than the revolution time of an electron in the first Bohr orbit of atomic hydrogen. The use of rotational Raman transitions, instead of vibrational transitions, is promising. For example, the $v' = 0$, $J' = 2 \rightarrow v'' = 0$, $J'' = 0$ rotational transition in H_2 will produce subfemtosecond pulses with a pulse spacing of 94 fs [11].

This work was supported by the U.S. Army Research Office, the U.S. Air Force Office of Scientific Research, and the U.S. Office of Naval Research.

[1] S. E. Harris and A. V. Sokolov, Phys. Rev. A **55**, R4019 (1997).
[2] K. Hakuta, M. Suzuki, M. Katsuragawa, and J. Z. Li, Phys. Rev. Lett. **79**, 209 (1997).
[3] H. Kawano, Y. Hirakawa, and T. Imasaka, IEEE J. Quantum Electron. **34**, 260 (1998).
[4] V. S. Butylkin, A. E. Kaplan, Yu. G. Khronopulo, and E. I. Yakubovich, *Resonant Nonlinear Interactions of Light with Matter* (Springer-Verlag, New York, 1977).
[5] A. E. Kaplan, Phys. Rev. Lett. **73**, 1243 (1994); A. E. Kaplan and P. L. Shkolnikov, J. Opt. Soc. Am. B **13**, 347 (1996).
[6] P. Antoine, A. L. 'Huiller, and M. Lewenstein, Phys. Rev. Lett. **77**, 1234 (1996); K. J. Schafer and K. C. Kulander, Phys. Rev. Lett. **78**, 638 (1997); S. E. Harris, J. J. Macklin, and T. W. Hansch, Opt. Commun. **100**, 487 (1993).
[7] M. O. Scully and M. S. Zubairy, *Quantum Optics* (Cambridge University Press, Cambridge, England, 1997); S. E. Harris, Phys. Today **50**, 36 (1997).
[8] O. Kocharovskaya and P. Mandel, Quantum Opt. **6**, 217 (1994); T. Ya. Popova, A. K. Popov, S. G. Rautian, and R. I. Sokolovskij, Sov. Phys. JETP **30**, 466 (1970).
[9] M. Jain, H. Xia, G. Y. Yin, A. J. Merriam, and S. E. Harris, Phys. Rev. Lett. **77**, 4326 (1996).
[10] A. C. Allison and A. Dalgarno, At. Data **1**, 289 (1970); G. Herzberg, *Molecular Spectra and Molecular Structure. I. Spectra of Dyatomic Molecules* (Van Nostrand Reinhold, New York, 1950).
[11] A. V. Sokolov, D. D. Yavuz, and S. E. Harris (to be published).

Light speed reduction to 17 metres per second in an ultracold atomic gas

Lene Vestergaard Hau*†, S. E. Harris‡, Zachary Dutton*† & Cyrus H. Behroozi*§

* *Rowland Institute for Science, 100 Edwin H. Land Boulevard, Cambridge, Massachusetts 02142, USA*
† *Department of Physics, § Division of Engineering and Applied Sciences, Harvard University, Cambridge, Massachusetts 02138, USA*
‡ *Edward L. Ginzton Laboratory, Stanford University, Stanford, California 94305, USA*

Techniques that use quantum interference effects are being actively investigated to manipulate the optical properties of quantum systems[1]. One such example is electromagnetically induced transparency, a quantum effect that permits the propagation of light pulses through an otherwise opaque medium[2–5]. Here we report an experimental demonstration of electromagnetically induced transparency in an ultracold gas of sodium atoms, in which the optical pulses propagate at twenty million times slower than the speed of light in a vacuum. The gas is cooled to nanokelvin temperatures by laser and evaporative cooling[6–10]. The quantum interference controlling the optical properties of the medium is set up by a 'coupling' laser beam propagating at a right angle to the pulsed 'probe' beam. At nanokelvin temperatures, the variation of refractive index with probe frequency can be made very steep. In conjunction with the high atomic density,

this results in the exceptionally low light speeds observed. By cooling the cloud below the transition temperature for Bose–Einstein condensation[11–13] (causing a macroscopic population of alkali atoms in the quantum ground state of the confining potential), we observe even lower pulse propagation velocities (17 m s^{-1}) owing to the increased atom density. We report an inferred nonlinear refractive index of 0.18 cm^2 W^{-1} and find that the system shows exceptionally large optical nonlinearities, which are of potential fundamental and technological interest for quantum optics.

The experiment is performed with a gas of sodium atoms cooled to nanokelvin temperatures. Our atom cooling set-up is described in some detail in ref. 14. Atoms emitted from a 'candlestick' atomic beam source[15] are decelerated in a Zeeman slower and loaded into a magneto-optical trap. In a few seconds we collect a cloud of 10^{10} atoms at a temperature of 1 mK and a density of 6×10^{11} cm^{-3}. The atoms are then polarization gradient cooled for a few milliseconds to 50 μK and optically pumped into the $F = 1$ ground state with an equal population of the three magnetic sublevels. We then turn all laser beams off and confine the atoms magnetically in the '4 Dee' trap[14]. Only atoms in the $M_F = -1$ state, with magnetic dipole moments directed opposite to the magnetic field direction (picked as the quantization axis), are trapped in the asymmetric harmonic trapping potential. This magnetic filtering results in a sample of atoms that are all in a single atomic state (state $|1\rangle$ in Fig. 1b) which allows adiabatic optical preparation of the atoms, as described below, and minimal heating of the cloud.

Next we evaporatively cool the atoms for 38 s to the transition temperature for Bose–Einstein condensation, T_c. The magnetic fields are then adjusted to adiabatically soften the trap. The resulting trapping potential has a frequency of $f_z = 21$ Hz along the symmetry (z) axis of the 4 Dee trap, and transverse frequencies $f_x = f_y = 69$ Hz. The bias field, parallel to the z axis, is 11 G. When we cool well below T_c, we are left with 1–2 million atoms in the condensate. For these parameters the transition occurs at a temperature of $T_c = 435$ nK and a peak density in the cloud of 5×10^{12} cm^{-3}.

We now apply a linearly polarized laser beam, the coupling beam, tuned to the transition between the unpopulated hyperfine states $|2\rangle$ and $|3\rangle$ (Fig. 1b). This beam couples states $|2\rangle$ and $|3\rangle$ and creates a quantum interference for a weaker probe laser beam (left circularly polarized) which is tuned to the $|1\rangle \rightarrow |3\rangle$ transition. A stable eigenstate (the 'dark state') of the atom in the presence of coupling and probe lasers is a coherent superposition of the two hyperfine ground states $|1\rangle$ and $|2\rangle$. The ratio of the probability amplitudes is such that the contributions to the atomic dipole moment induced by the two lasers exactly cancel. The quantum interference occurs in a narrow interval of probe frequencies, with a width determined by the coupling laser power.

Figure 2a shows the calculated transmission of the probe beam as a function of its detuning from resonance for parameters which are typical of this work. In the absence of dephasing of the $|1\rangle \rightarrow |2\rangle$ transition, the quantum interference would be perfect, and at line centre, the transmission would be unity. Figure 2b shows the refractive index for the probe beam as a function of detuning. Due to the very small Doppler broadening of the $|1\rangle \rightarrow |2\rangle$ transition in our nanokelvin samples, application of very low coupling intensity leads to a transparency peak with a width much smaller than the natural line width of the $|1\rangle \rightarrow |3\rangle$ transition. Correspondingly, the dispersion curve is much steeper than can be obtained by any other technique, and this results in the unprecedented low group velocities reported here. The group velocity v_g for a propagating electromagnetic pulse is[16–19]:

$$v_g = \frac{c}{n(\omega_p) + \omega_p \dfrac{dn}{d\omega_p}} \approx \frac{hc\epsilon_0}{2\omega_p} \frac{|\Omega_c|^2}{|\mu_{13}|^2 N} \qquad (1)$$

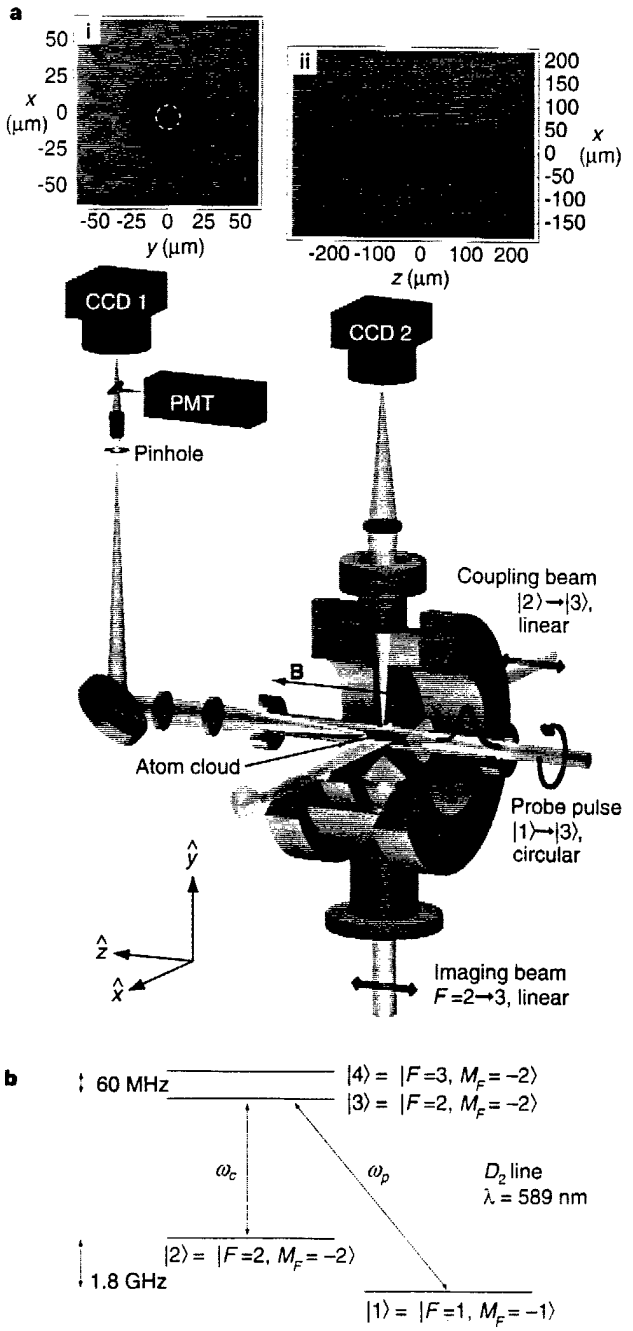


Figure 1 Experimental set-up. A 'coupling' laser beam propagates along the x axis with its linear polarization along the 11-G bias field in the z direction. The 'probe' laser pulse propagates along the z axis and is left-circularly polarized. With a flipper mirror in front of the camera CCD 1, we direct this probe beam either to the camera or to the photomultiplier (PMT). For pulse delay measurements, we place a pinhole in an external image plane of the imaging optics and select a small area, 15 μm in diameter, of the probe beam centred on the atom clouds (as indicated by the dashed circle in inset (i)). The pulse delays are measured with the PMT. The imaging beam propagating along the y axis is used to image atom clouds onto camera CCD 2 to find the length of the clouds along the pulse propagation direction (z axis) for determination of light speeds. Inset (ii) shows atoms cooled to 450 nK which is 15 nK above T_c. (Note that this imaging beam is never applied at the same time as the probe pulse and coupling laser). The position of a cloud and its diameter in the two transverse directions, x and y, are found with CCD 1. Inset (i) shows an image of a condensate.

Here $n(\omega_p)$ is the refractive index at probe frequency ω_p (rad s^{-1}), $|\Omega_c|^2$ is the square of the Rabi frequency for the coupling laser and varies linearly with intensity, μ_{13} is the electric dipole matrix element between states $|1\rangle$ and $|3\rangle$, N is the atomic density, and ϵ_0 is the permittivity of free space. At line centre, the refractive index is unity, and the second term in the denominator of equation (1) dominates the first. An important characteristic of the refractive index profile is that on resonance the dispersion of the group velocity is zero (see ref. 16), that is, $d^2n/d\omega_p^2 = 0$, and to lowest order, the pulse maintains its shape as it propagates. The established quantum interference allows pulse transmission through our atom clouds which would otherwise have transmission coefficients of e^{-110} (below T_c), and creates a steep dispersive profile and very low group velocity for light pulses propagating through the clouds.

We note that the centres of the curves in Fig. 2 are shifted by 0.6 MHz from probe resonance. This is due to a coupling of state $|2\rangle$ to state $|4\rangle$ through the coupling laser field, which results in an a.c. Stark shift of level $|2\rangle$ and a corresponding line shift of the $2 \rightarrow 3$ transition. As the transparency peak and unity refractive index are obtained at two-photon resonance, this leads to a refractive index at the $1 \rightarrow 3$ resonance frequency which is different from unity. The difference is proportional to the a.c. Stark shift and hence to the coupling laser intensity, which is important for predicting the nonlinear refractive index as described below.

A diagram of the experiment is shown in Fig. 1a. The 2.5-mm-diameter coupling beam propagates along the x axis with its linear polarization parallel to the **B** field. The 0.5-mm-diameter, σ^- polarized probe beam propagates along the z axis. The size and position of the atom cloud in the transverse directions, x and y, are obtained by imaging the transmission profile of the probe beam after the cloud onto a charge-coupled-device (CCD) camera. An image of a condensate is shown as inset (i). A 55 mW cm^{-2} coupling laser beam was present during the 10-μs exposure of the atoms to a 5 mW cm^{-2} probe beam tuned close to resonance. The $f/7$ imaging optics are diffraction-limited to a resolution of 7 μm.

During the pulse delay experiments, a pinhole (placed in an external image plane of the lens system) is used to select only the part of the probe light that has passed through the central 15 μm of the atom cloud where the column density is the greatest. The outline of the pinhole is indicated with the dashed circle in inset (i).

Both coupling and probe beams are derived from the same dye laser. The frequency of the coupling beam is set by an acousto-optic modulator (AOM) to the $|2\rangle \rightarrow |3\rangle$ resonance. Here we take into account both Zeeman shifts and the a.c. Stark shift described above.

The corresponding probe resonance is found by measuring the transmission of the probe beam as a function of its frequency. We apply a fast frequency sweep, across 32 MHz in 50 μs, and determine resonance from the transmission peak. The sweep is controlled by a separate AOM. The frequency is then fixed at resonance, and the temporal shape of the probe pulse is generated by controlling the r.f. drive power to the AOM. The resulting pulse is approximately gaussian with a full-width at half-maximum of 2.5 μs. The peak power is 1 mW cm^{-2} corresponding to a Rabi frequency of $\Omega_p = 0.20A$, where the Einstein A coefficient is 6.3×10^7 rad s^{-1}. To avoid distortion of the pulse, it is made of sufficient duration that its Fourier components are contained within the transparency peak.

Probe pulses are launched along the z axis 4 μs after the coupling beam is turned on (the coupling field is left on for 100 μs). Due to the magnetic filtering discussed above, all atoms are initially in state $|1\rangle$ which is a dark state in the presence of the coupling laser only. When the pulse arrives, the atoms adiabatically evolve so that the probability amplitude of state $|2\rangle$ is equal to the ratio $\Omega_p/(\Omega_p^2 + \Omega_c^2)^{1/2}$, where Ω_p is the probe Rabi frequency. To establish the coherent superposition state, energy is transferred from the

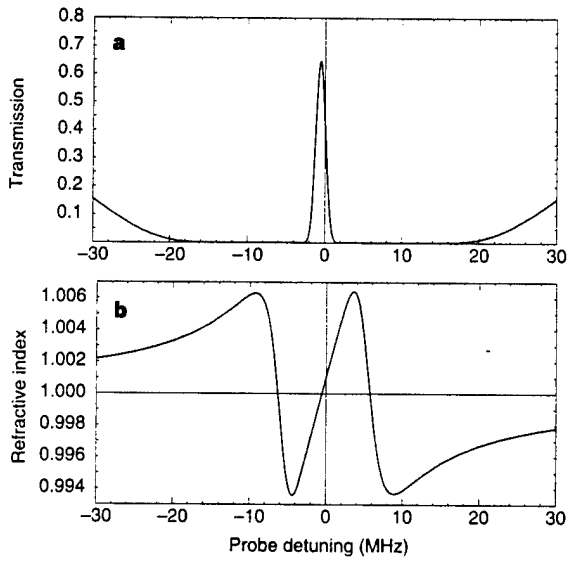


Figure 2 Effect of probe detuning. **a**, Transmission profile. Calculated probe transmission as a function of detuning from the $|1\rangle \rightarrow |3\rangle$ resonance for an atom cloud cooled to 450 nK, with a peak density of 3.3×10^{12} cm^{-3} and a length of 229 μm (corresponding to the cloud in inset (ii) of Fig. 1a). The coupling laser is resonant with the $|2\rangle \rightarrow |3\rangle$ transition and has a power density of 52 mW cm^{-2}. **b**, Refractive index profile. The calculated refractive index is shown as a function of probe detuning for the same parameters as in **a**. The steepness of the slope at resonance is inversely proportional to the group velocity of transmitted light pulses and is controlled by the coupling laser intensity. Note that as a result of the a.c. Stark shift of the $|2\rangle \rightarrow |3\rangle$ transition, caused by a coupling of states $|2\rangle$ and $|4\rangle$ through the coupling laser field, the centre of the transmission and refractive index profiles is shifted by 0.6 MHz. The shift of the refractive index profile results in the nonlinear refractive index described in the text.

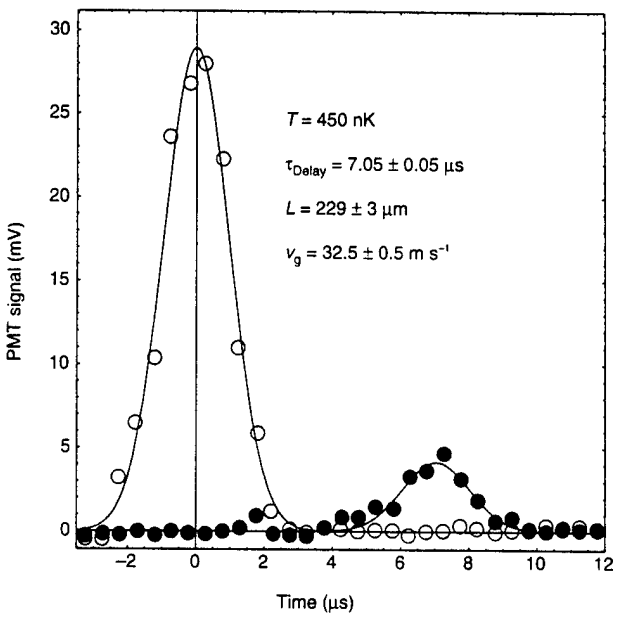


Figure 3 Pulse delay measurement. The front pulse (open circles) is a reference pulse with no atoms in the system. The other pulse (filled circles) is delayed by 7.05 μs in a 229-μm-long atom cloud (see inset (ii) in Fig. 1a). The corresponding light speed is 32.5 m s^{-1}. The curves represent gaussian fits to the measured pulses.

front of the probe pulse to the atoms and the coupling laser field. At the end of the pulse, the atoms adiabatically return to the original state $|1\rangle$ and the energy returns to the back of the probe pulse with no net energy and momentum transfer to the atomic cloud. Because the refractive index is unity, the electric field is unchanged as the probe pulse enters the medium. As the group velocity is decreased, the total energy density must increase so as to keep constant the power per area. This increase is represented by the energy stored in the atoms and the coupling laser field during pulse propagation through the cloud.

The pulses are recorded with a photomultiplier (3-ns response time) after they penetrate the atom clouds. The output from the photomultiplier is amplified by a 150-MHz-bandwidth amplifier and the waveforms are recorded on a digital scope. With a 'flipper' mirror in front of the camera we control whether the probe beam is directed to the camera or to the photomultiplier.

The result of a pulse delay measurement is shown in Fig. 3. The front pulse is a reference pulse obtained with no atoms present. The pulse delayed by 7.05 μs was slowed down in an atom cloud with a length of 229 μm (see Fig. 1a, inset (ii)). The resulting light speed is 32.5 m s^{-1}. We used a coupling laser intensity of 12 mW cm^{-2} corresponding to a Rabi frequency of $\Omega_c = 0.56A$. The cloud was cooled to 450 nK (which is 15 nK above T_c), the peak density was 3.3×10^{12} cm^{-3}, and the total number of atoms was 3.8×10^6. From these numbers we calculate that the pulse transmission coefficient would be e^{-63} in the absence of the coupling laser. The probe pulse was indeed observed to be totally absorbed by the atoms when the coupling beam was left off. Inhomogeneous broadening due to spatially varying Zeeman shifts is negligible (~20 kHz) for the low temperatures and correspondingly small cloud sizes used here.

The size of the atom cloud in the z direction is obtained with another CCD camera. For this purpose, we use a separate 1 mW cm^{-2} laser beam propagating along the vertical y axis and tuned 20 MHz below the $F = 2 \rightarrow 3$ transition. The atoms are pumped to the $F = 2$ ground state for 10 μs before the imaging which is performed with an exposure time of 10 μs. We image the transmission profile of the laser beam after the atom cloud with diffraction-limited $f/5$ optics. An example is shown in Fig. 1a, inset (ii), where the asymmetry of the trap is clear from the cloud's elliptical profile. We note that the imaging laser is never applied at the same time as the coupling laser and probe pulse, and for each recorded pulse or CCD picture a new cloud is loaded.

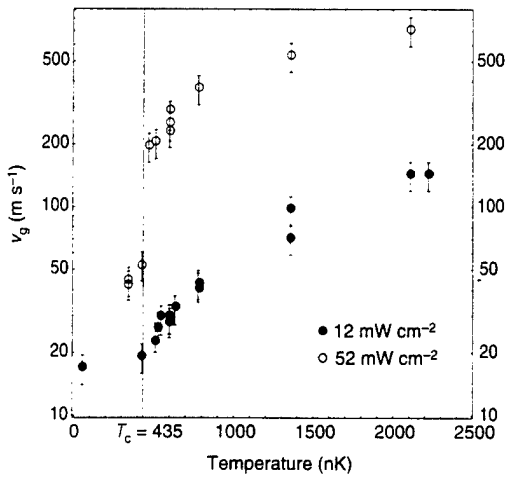


Figure 4 Light speed versus atom cloud temperature. The speed decreases with temperature due to the atom density increase. The open circles are for a coupling power of 52 mW cm^{-2} and the filled circles are for a coupling power of 12 mW cm^{-2}. The temperature T_c marks the transition temperature for Bose-Einstein condensation. The decrease in group velocity below T_c is due to a density increase of the atom cloud when the condensate is formed. From imaging measurements we obtain a maximum atom density of 8×10^{13} cm^{-3} at a temperature of 200 nK. Here, the dense condensate component constitutes 60% of all atoms, and the total atom density is 16 times larger than the density of a non-condensed cloud at T_c. The light speed measurement at 50 nK is for a cloud with a condensate fraction ≥90%. The finite dephasing rate due to state $|4\rangle$ does not allow pulse penetration of the most dense clouds. This problem could be overcome by tuning the laser to the D_1 line as described in the text.

We measured a series of pulse delays and corresponding cloud sizes for atoms cooled to temperatures between 2.5 μK and 50 nK. From these pairs of numbers we obtain the corresponding propagation velocities (Fig. 4). The open circles are for a coupling power of 52 mW cm^{-2} ($\Omega_c = 1.2A$). The light speed is inversely proportional to the atom density (equation (1)) which increases with lower temperatures, with an additional density increase when a condensate is formed. The filled circles are for a coupling power of 12 mW cm^{-2}. The lower coupling power is seen to cause a decrease of group velocities in agreement with equation (1). We obtain a light speed of 17 m s^{-1} for pulse propagation in an atom cloud initially prepared as an almost pure Bose–Einstein condensate (condensate fraction is ≥90%). Whether the cloud remains a condensate during and after pulse propagation is an issue that is beyond the scope of this Letter.

Transitions from state $|2\rangle$ to state $|4\rangle$, induced by the coupling laser (detuned by 60 MHz from this transition), result in a finite decay rate of the established coherence between states $|1\rangle$ and $|2\rangle$ and limit pulse transmission. The dephasing rate is proportional to the power density of the coupling laser and we expect, and find, that probe pulses have a peak transmission that is independent of coupling intensity and a velocity which reduces linearly with this intensity. The dephasing time is determined from the slope of a semi-log plot of transmission versus pulse delay[19]. At a coupling power of 12 mW cm^{-2}, we measured a dephasing time of 9 μs for atom clouds just above T_c.

Giant Kerr nonlinearities are of interest for areas of quantum optics such as optical squeezing, quantum nondemolition, and studies of nonlocality. It was recently proposed that they may be obtained using electromagnetically induced transparency[20]. Here we report the first (to our knowledge) measurement of such a nonlinearity. The refractive index for zero probe detuning is given by $n = 1 + (n_2 I_c)$ where I_c is the coupling laser intensity, and n_2 the cross phase nonlinear refractive index. As seen from Fig. 2b, the nonlinear term $(n_2 I_c)$ equals the product of the slope of the refractive index at probe resonance and the a.c. Stark shift of the $|2\rangle \rightarrow |3\rangle$ transition caused by the coupling laser. We can then express n_2 by the formula (see equation (1));

$$n_2 = \frac{\Delta\omega_S}{I_c}\frac{dn}{d\omega_p} \approx \frac{1}{2\pi}\frac{\Delta\omega_S}{I_c}\frac{\lambda}{v_g} \qquad (2)$$

where $\Delta\omega_S$ is the a.c. Stark shift, proportional to I_c, and λ the wavelength of the probe transition. We measured an a.c. Stark shift of 1.3×10^6 rad s^{-1} for a coupling laser intensity of 40 mW cm^{-2}. For a measured group velocity of 17 m s^{-1} (Fig. 4), we obtain a nonlinear refractive index of 0.18 cm^2 W^{-1}. This nonlinear index is ~10^6 times greater than that measured in cold Cs atoms[21].

With a system that avoids the $|1\rangle$-$|2\rangle$ dephasing rate described above (which can be obtained by tuning to the D_1 line in sodium), the method used here could be developed to yield the collision-induced dephasing rate of the double condensate which is generated in the process of establishing electromagnetically induced transparency (see also refs 22, 23). In that case, the square of the probability amplitude for state $|3\rangle$ could be kept below 10^{-5} during pulse propagation, with no heating of the condensate as a result. With improved frequency stability of our set-up and lower coupling intensities, even lower light speeds would be possible, perhaps of the order of centimetres per second, comparable to the speed of

sound in a Bose–Einstein condensate. Under these conditions we expect phonon excitation during light pulse propagation through the condensate. By deliberately tuning another laser beam to the $|2\rangle \rightarrow |4\rangle$ transition, it should be possible to demonstrate optical switching at the single photon level[24]. Finally, we note that during propagation of the atom clouds, light pulses are compressed in the z direction by a ratio of c/v_g. For our experimental parameters, that results in pulses with a spatial extent of only 43 μm. □

Received 3 November; accepted 21 December 1998.

1. Knight, P. L., Stoicheff, B. & Walls, D. (eds) Highlights in quantum optics. *Phil. Trans. R. Soc. Lond. A* **355**, 2215–2416 (1997).
2. Harris, S. E. Electromagnetically induced transparency. *Phys. Today* **50(7)**, 36–42 (1997).
3. Scully, M. O. & Zubairy, M. S. *Quantum Optics* (Cambridge Univ. Press, 1997).
4. Arimondo, E. in *Progress in Optics* (ed. Wolf, E.) 257–354 (Elsevier Science, Amsterdam, 1996).
5. Bergmann, K., Theuer, H. & Shore, B. W. Coherent population transfer among quantum states of atoms and molecules. *Rev. Mod. Phys.* **70**, 1003–1006 (1998).
6. Chu, S. The manipulation of neutral particles. *Rev. Mod. Phys.* **70**, 685–706 (1998).
7. Cohen-Tannoudjii, C. N. Manipulating atoms with photons. *Rev. Mod. Phys.* **70**, 707–719 (1998).
8. Phillips, W. D. Laser cooling and trapping of neutral atoms. *Rev. Mod. Phys.* **70**, 721–741 (1998).
9. Hess, H. F. Evaporative cooling of magnetically trapped and compressed spin-polarized hydrogen. *Phys. Rev. B* **34**, 3476–3479 (1986).
10. Masuhara, N. *et al.* Evaporative cooling of spin-polarized atomic hydrogen. *Phys. Rev. Lett.* **61**, 935–938 (1988).
11. Anderson, M. H., Ensher, J. R., Matthews, M. R., Wieman, C. E. & Cornell, E. A. Observation of Bose-Einstein condensation in a dilute atomic vapor. *Science* **269**, 198–201 (1995).
12. Davis, K. B. *et al.* Bose-Einstein condensation in a gas of sodium atoms. *Phys. Rev. Lett.* **75**, 3969–3973 (1995).
13. Bradley, C. C., Sackett, C. A. & Hulet, R. G. Bose-Einstein condensation of lithium: observation of limited condensate number. *Phys. Rev. Lett.* **78**, 985–989 (1997).
14. Hau, L. V. *et al.* Near-resonant spatial images of confined Bose-Einstein condensates in a 4-Dee magnetic bottle. *Phys. Rev. A* **58**, R54–R57 (1998).
15. Hau, L. V., Golovchenko, J. A. & Burns, M. M. A new atomic beam source: The "candlestick". *Rev. Sci. Instrum.* **65**, 3746–3750 (1994).
16. Harris, S. E., Field, J. E. & Kasapi, A. Dispersive properties of electromagnetically induced transparency. *Phys. Rev. A* **46**, R29–R32 (1992).
17. Grobe, R., Hioe, F. T. & Eberly, J. H. Formation of shape-preserving pulses in a nonlinear adiabatically integrable system. *Phys. Rev. Lett.* **73**, 3183–3186 (1994).
18. Xiao, M., Li, Y.-Q., Jin, S.-Z. & Gea-Banacloche, J. Measurement of dispersive properties of electromagnetically induced transparency in rubidium atoms. *Phys. Rev. Lett.* **74**, 666–669 (1995).
19. Kasapi, A., Jain, M., Yin, G. Y. & Harris, S. E. Electromagnetically induced transparency: propagation dynamics. *Phys. Rev. Lett.* **74**, 2447–2450 (1995).
20. Schmidt, H. & Imamoglu, A. Giant Kerr nonlinearities obtained by electromagnetically induced transparency. *Opt. Lett.* **21**, 1936–1938 (1996).
21. Lambrecht, A., Courty, J. M., Reynaud, S. & Giacobino, E. Cold atoms: A new medium for quantum optics. *Appl. Phys. B* **60**, 129–134 (1995).
22. Hall, D. S., Matthews, M. R., Wieman, C. E. & Cornell, E. A. Measurements of relative phase in two-component Bose-Einstein condensates. *Phys. Rev. Lett.* **81**, 1543–1546 (1998).
23. Ruostekoski, J. & Walls, D. F. Coherent population trapping of Bose-Einstein condensates: detection of phase diffusion. *Eur. Phys. J. D* (submitted).
24. Harris, S. E. & Yamamoto, Y. Photon switching by quantum interference. *Phys. Rev. Lett.* **81**, 3611–3614 (1998).

Acknowledgements. We thank J. A. Golovchenko for discussions and C. Liu for experimental assistance. L.V.H. acknowledges support from the Rowland Institute for Science. S.E.H. is supported by the US Air Force Office of Scientific Research, the US Army Research Office, and the US Office of Naval Research. C.H.B. is supported by an NSF fellowship.

Correspondence and requests for materials should be addressed to L.V.H. (e-mail: hau@rowland.org).

Efficient Nonlinear Frequency Conversion in an All-Resonant Double-Λ System

Andrew J. Merriam, S. J. Sharpe, M. Shverdin, D. Manuszak, G. Y. Yin, and S. E. Harris
Edward L. Ginzton Laboratory, Stanford University, Stanford, California 94305
(Received 3 February 2000)

We demonstrate efficient, pulsed, gas-phase, nonlinear frequency conversion in a quadruply resonant, double-Λ system and, simultaneously, verify theoretical predictions of Rabi-frequency matching unique to absorbing nonlinear media. This system is used to up-convert ultraviolet light at 233 nm to the vacuum ultraviolet at 186 nm in atomic Pb vapor with small-signal conversion efficiencies exceeding 30% and with modest atomic density-length (NL) products (scale 10^{14} cm^{-2}) and optical power densities (10–100 kW/cm^2).

PACS numbers: 42.50.Gy, 42.50.Hz, 42.65.Ky, 42.79.Nv

It has been shown that the techniques of electromagnetically induced transparency (EIT) may greatly improve the efficiency of gas-phase nonlinear optical processes [1,2]. EIT facilitates the preparation of near-maximal coherence of a dipole-forbidden transition without absorption or self-phase modulation of the applied fields; the phase-coherent atomic ensemble thus created acts as a strong local oscillator which may mix with the spectral components of the applied fields to generate a corresponding spectrum of sum or difference frequencies. Previous work in both the gas [1,2] and solid [3] phases has focused on the reactive case (i.e., large detuning of the generated frequency from any resonances and near-lossless propagation) and has demonstrated efficient parametric frequency conversion to the ultraviolet and vacuum ultraviolet (VUV).

In this Letter, we explore gas-phase, nonlinear frequency up-conversion in a quadruply resonant atomic system. This atom-field combination is known as a resonant double-Λ configuration and has been studied in the context of EIT [4], lasers without inversion [5], and nonlinear optics [6]. The short-wavelength up-converted radiation is generated directly on resonance under conditions where, in the absence of the other fields, it would be strongly absorbed. We demonstrate, however, that through the up-conversion process, the atomic system renders itself transparent to all four fields. Transparency is achieved once the complex ratio of Rabi frequencies in each Λ-channel is equal and, once transparent, no further changes to the relative phase or amplitude of each field occur [7]. Additionally, we observe important saturation mechanisms which may limit the generated radiation intensity attainable with this type of frequency converter.

We begin with a brief theoretical discussion of this work. We consider a four-state system in the rotating wave approximation, as shown in the inset in Fig. 1. The coupling laser, tuned to the $|2\rangle \rightarrow |3\rangle$ transition with Rabi frequency Ω_c (406 nm), and the probe laser, tuned to the $|1\rangle \rightarrow |3\rangle$ transition with Rabi frequency Ω_p (283 nm), adiabatically establish a large atomic coherence of the Raman transition, which is described by the off-diagonal density matrix element ρ_{12} [2]. A third laser field with radian frequency ω_e is tuned to resonance with the $|2\rangle \rightarrow |4\rangle$ transition and mixes with the coherence to generate a fourth field at ω_h resonant with the $|1\rangle \rightarrow |4\rangle$ transition. The (small-signal) electric-field attenuation coefficients of the mixing and generated fields are $\alpha_e = N\sigma_e\rho_{22}/2$ and $\alpha_h = N\sigma_h\rho_{11}/2$, where N is the atom density, ρ_{jj} is the total fraction of atoms in state $|j\rangle$, and σ_e and σ_h are the single-atom, Doppler-broadened, resonant, absorption cross sections. The coherently trapped population is distributed according to $\rho_{11} = \Omega_c^2/(\Omega_c^2 + \Omega_p^2)$, $\rho_{22} = \Omega_p^2/(\Omega_c^2 + \Omega_p^2)$, and $\rho_{12} = -\Omega_p\Omega_c^*/(\Omega_p^2 + \Omega_c^2)$. With these definitions, in a plane-wave approximation and ignoring saturation effects and light shifts, the propagation equations for a quadruply resonant frequency converter from Ω_e (233 nm) to Ω_h (186 nm) are [2]

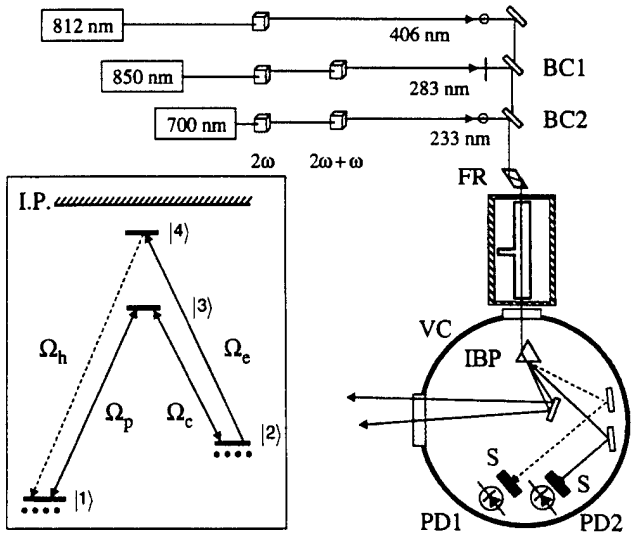


FIG. 1. Schematic diagram of the experimental apparatus. BC1, etc., described in main text. Inset: Energy-level diagram of the double-Λ system in ^{208}Pb vapor. Laser fields with Rabi frequencies Ω_p, Ω_c, and Ω_e are applied, and Ω_h is generated. State designations and energies: $|1\rangle$, $6p^2\ ^3P_0$, 0.0 cm^{-1} (ground); $|2\rangle$, $6p^2\ ^3P_2$, 10 650.3271 cm^{-1}; $|3\rangle$, $6p7s\ ^3P_1$, 35 287.2244 cm^{-1}; $|4\rangle$, $6p9s\ ^3P_1$, 53 511.1485 cm^{-1}. The ionization potential of Pb I is 59 819 cm^{-1} (7.42 eV).

 0031-9007/00/84(23)/5308(4)$15.00

$$\frac{\partial \Omega_e}{\partial z} + \alpha_e \Omega_e = -\frac{N\sigma_e}{2} \rho_{12}^* \Omega_h \exp(+i\Delta k z), \quad (1a)$$

$$\frac{\partial \Omega_h}{\partial z} + \alpha_h \Omega_h = -\frac{N\sigma_h}{2} \rho_{12} \Omega_e \exp(-i\Delta k z). \quad (1b)$$

Since k-vector matching of the interacting laser fields is automatically satisfied by resonant copropagating fields in the rotating wave approximation, $\Delta k = 0$. The solutions of Eqs. (1) with boundary conditions $\Omega_h(z = 0) = 0$ and $\Omega_e(z = 0) = \Omega_e(0)$ are

$$\frac{\Omega_h(z)}{\Omega_e(0)} = iN\sigma_h\rho_{12} \exp\left[-\frac{\alpha_T z}{2}\right] \frac{\sin(i\alpha_T z/2)}{\alpha_T}$$

$$= \left(\frac{\Omega_p}{\Omega_c}\right)\left(\frac{\alpha_h}{\alpha_T}\right)(1 - e^{-\alpha_T z}), \quad (2a)$$

$$\frac{\Omega_e(z)}{\Omega_e(0)} = \frac{\alpha_h}{\alpha_T} + \frac{\alpha_e}{\alpha_T} e^{-\alpha_T z}, \quad (2b)$$

where $\alpha_T = \alpha_e + \alpha_h$. This solution assumes that the atomic coherence ρ_{12} established by the fields in the first Λ-channel is unaffected by the generation process; this is ensured by maintaining $\Omega_e(0)$ an order of magnitude less than Ω_p and Ω_c during the experiment.

The central result of this work is that, in the limit of many absorption lengths ($\alpha_T z \to \infty$), the nonlinear generation process matches the ratio of Rabi frequencies in each Λ-channel so that $\Omega_h(z)/\Omega_e(z) \to \Omega_p/\Omega_c$. The frequency conversion behavior predicted by Eq. (2) is dramatically different from that witnessed in earlier, far-detuned experiments [1]; no longer is there parametric power oscillation between fields as a function of distance into the nonlinear medium, but, rather, monotonic growth towards an asymptotic value. This result may be viewed as a special case of matched pulses [7] in a double-Λ system; atoms evolved into the coherent population-trapped eigenstate are invisible to any pair of optical frequency fields on the $|1\rangle \leftrightarrow |4\rangle$ and $|2\rangle \leftrightarrow |4\rangle$ transitions whose complex ratio of Rabi frequencies is Ω_p/Ω_c. Pairs of input fields which differ from this ratio undergo selective absorption of unmatched Fourier components and nonlinear generation until they conform to this ratio and are thereby decoupled from the medium. This behavior is quite different from the density invariance of previous resonant frequency conversion experiments caused by competition between four-wave mixing and two-photon absorption [8].

We now turn to a description of the experiment. A schematic diagram of the experimental setup is shown in Fig. 1. Three linearly polarized, coherent beams at 406, 283, and 233 nm are spatially and temporally overlapped by beam combiners BC1 and BC2, converted to circular polarization by a Fresnel rhomb FR, and propagate collinearly through the Pb atomic vapor contained in a heated sidearm cell. The output beams propagate ~1 cm in atmosphere into a vacuum chamber VC (maintained <500 mTorr) and are dispersed by an isosceles Brewster prism IBP. The generated field at 186 nm and residual mixing field at 233 nm are directed onto sodium salicylate scintillators S. The fluorescence signals are sampled by fast photodiodes PD1 and PD2. Each waveform is digitized and downloaded to a computer. The data are analyzed, sorted by the pulse timing, and plotted; data with pulse timing overlap errors less than 5 ns are retained. The data shown in Figs. 2–5 are from individual events and are not averaged.

The laser setup is similar to that used in previous experiments [2,9]. All beams are collimated with intensity FWHM beam diameters of 2.0 mm (coupling laser, 406 nm, 30-ns pulse width), 2.12 mm (probe laser, 283 nm, 15-ns pulse width), and 1.24 mm (mixing laser, 233 nm, 15-ns pulse width). The area of the generated beam is inferred to be the same as that of the mixing laser. Laser frequencies are determined using a Burleigh Model 4500 pulsed wave meter with a maximum resolution of 0.01 cm^{-1} (300 MHz). Typical energies and (peak) Rabi frequencies for the probe, coupling, mixing, and generated lasers are 50 μJ (0.25 cm^{-1}), 50 μJ (0.30 cm^{-1}), 1 μJ (0.0148 cm^{-1}), and 0.1 μJ (5.31×10^{-3} cm^{-1}), respectively. (These beam energies are a factor of 200 lower than those employed in previous EIT-enhanced nonlinear optics experiments [1,2].)

The 186-nm vacuum ultraviolet radiation is generated as a collimated beam which propagates collinearly (i.e., on axis) with the three applied fields. Typical pulse energies are less than 1 μJ and are determined using a scintillator/photodiode combination which has been calibrated with known 233-nm beam energies [10].

The atomic vapor is 99.86% isotopically enriched ^{208}Pb metal (Oak Ridge National Laboratory), heated within a 25-cm-long fused quartz sidearm cell manufactured by Opthos Instruments. As in previous work [11], densities

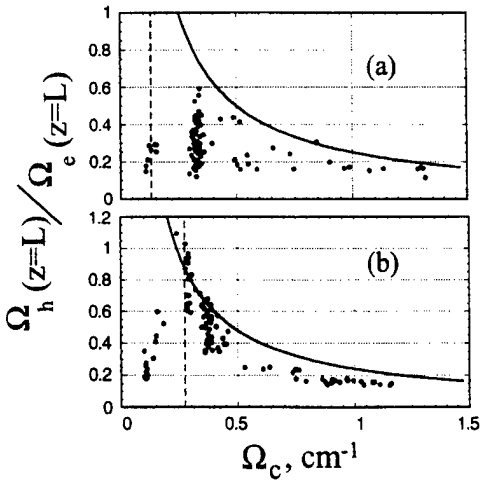


FIG. 2. Ratio of generated 186-nm and output 233-nm Rabi frequencies, at the exit of the cell $\Omega_h(z = L)/\Omega_e(z = L)$, as a function of the coupling laser Rabi frequency, at fixed Ω_p. (a) For NL product of 1.2×10^{14} cm^{-2}. (b) For NL product of 5.6×10^{14} cm^{-2}. The dashed vertical line in each figure is the value of Ω_c required for weak-probe EIT. The hyperbola is the $\alpha_T L \to \infty$ theoretical prediction of Eq. (2), taking Ω_p equal to (a) 0.25 cm^{-1} and (b) 0.24 cm^{-1}.

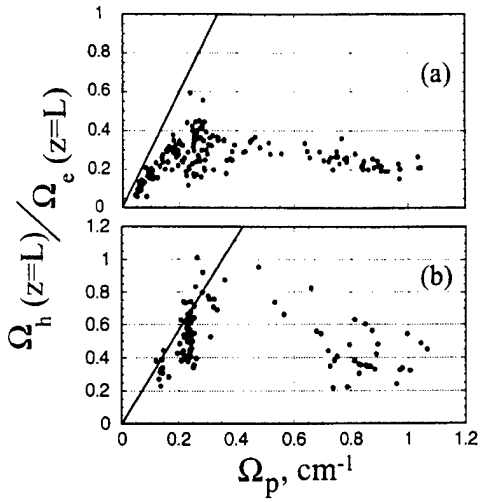


FIG. 3. Ratio of generated 186-nm and output 233-nm Rabi frequencies at the exit of the cell $\Omega_h(z = L)/\Omega_e(z = L)$, as a function of the probe laser Rabi frequency, at fixed Ω_c. (a) For NL product of 1.2×10^{14} cm^{-2}. (b) For NL product of 5.6×10^{14} cm^{-2}. The solid line is the $\alpha_T L \to \infty$ theoretical prediction, taking Ω_c equal to (a) 0.33 cm^{-1} and (b) 0.35 cm^{-1}.

are determined using the curve-of-growth method (to an accuracy of $\pm 10\%$). At 600 °C, the resonant absorption cross sections are $\sigma_e = 2.21 \times 10^{-13}$ cm^2 and $\sigma_h = 2.83 \times 10^{-13}$ cm^2. We investigate the behavior of the quadruply resonant, frequency converter at two density-length (NL) products: (a) 1.2×10^{14} cm^{-2} (588 °C) and (b) 5.6×10^{14} cm^{-2} (650 °C).

In Fig. 2, we study the dependence of the ratio of the Rabi frequencies of the generated VUV and output mixing fields at the exit of the sidearm cell, $\Omega_h(z = L)/\Omega_e(z = L)$, on the coupling laser Rabi frequency Ω_c. Here, the probe laser Rabi frequency is held at (a) 0.25 ± 0.01 cm^{-1} and (b) 0.24 ± 0.01 cm^{-1}, and $\Omega_e(0) < 0.025$ cm^{-1}. The curved, solid lines represent the theoretical prediction of Eqs. (2) at these values of Ω_p, assuming complete Rabi-frequency matching (i.e., taking $\alpha_T L \to \infty$). The dashed, vertical line indicates the coupling laser Rabi frequency at which, for each density, the step-up in EIT occurs (i.e., where, in the absence of the mixing laser, the transmission of a weak probe laser field through the cell exceeds 50%). Above this value, sufficient preparation energy has been applied to the system to ensure evolution of the ensemble into the population-trapped eigenstate [12]. In the following figures, only those points with $\Omega_c > 0.3$ cm^{-1} are shown.

In Fig. 3 we show $\Omega_h(z = L)/\Omega_e(z = L)$ as a function of the probe laser Rabi frequency Ω_p at fixed Ω_c and the $\alpha_T L \to \infty$ theoretical prediction. A striking feature of these data is the departure from theory when the Rabi frequency of the probe laser exceeds that of the coupling laser. This is most likely related to a failure of EIT; increasing the probe laser strength relative to the coupling laser in the presence of nonzero Raman-transition dephasing rates exacerbates two-photon absorption and causes non-negligible saturation and loss of coherence on time scales comparable to the pulse widths. (The dephasing time T_2 of the Raman transition coherence is ≈ 30 ns.) We expect that the use of shorter (but still adiabatic) pulses in such an experiment would limit this saturation and increase the strong-probe conversion efficiency. This strong-probe saturation behavior has been observed in other work in a continuous-wave regime [13]. We limit display of data in the remaining figures to those points where $\Omega_p < \Omega_c$.

In Fig. 4, we study the ratio of output and input mixing field Rabi frequencies $\Omega_e(z = L)/\Omega_e(0)$ as a function of

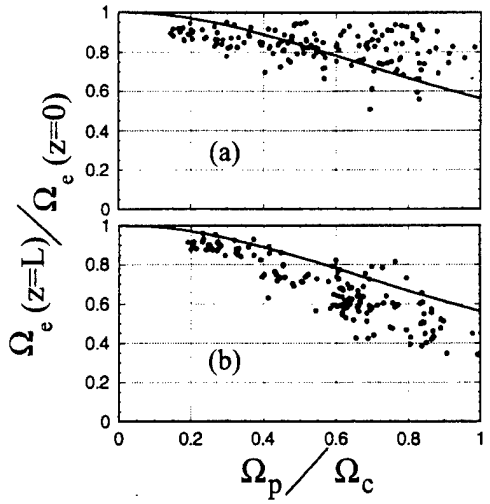


FIG. 4. Ratio of the output and input 233-nm Rabi frequencies $\Omega_e(z = L)/\Omega_e(z = 0)$ as a function of the ratio of probe and coupling laser Rabi frequencies Ω_p/Ω_c. (a) For NL product of 1.2×10^{14} cm^{-2}. (b) For NL product of 5.6×10^{14} cm^{-2}. The solid line is the $\alpha_T L \to \infty$ theoretical prediction.

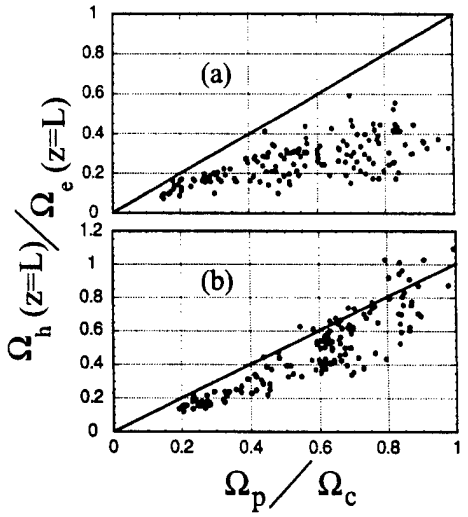


FIG. 5. Evolution to matched pulses. Ratio of generated 186-nm and output 233-nm Rabi frequencies at the exit of the cell $\Omega_h(z = L)/\Omega_e(z = L)$, as a function of the ratio of probe and coupling laser Rabi frequencies Ω_p/Ω_c. (a) For NL product of 1.2×10^{14} cm^{-2}. (b) For NL product of 5.6×10^{14} cm^{-2}. The solid line is the $\alpha_T L \to \infty$ theoretical prediction.

the ratio of the probe and coupling laser Rabi frequencies and compare with the predictions of Eq. (2b). Variation with the probe and coupling lasers is caused by the implicit dependence of the attenuation coefficients α_e and α_h on the population terms ρ_{22} and ρ_{11}. Note that, in the limit of weak probe (vanishing coherence), no population is transferred to state $|2\rangle$, and there is no appreciable absorption of the mixing field.

The data of the previous figures are combined in Fig. 5 to illustrate the evolution to a Rabi-frequency matching regime at the higher temperature. The opacity of the low density case ($\alpha_T L \approx 13$) should be more than sufficient to establish the Rabi-frequency matching condition, and no density dependence should remain. It is clear from the data, however, that matching is not attained until the higher density case ($\alpha_T L \approx 57$). The discrepancy arises from neglecting saturation and power broadening of the $|1\rangle \leftrightarrow |4\rangle$ and $|2\rangle \leftrightarrow |4\rangle$ transitions in the derivation of Eqs. (1). This assumption requires that the mixing and generated field Rabi frequencies Ω_e and Ω_h are both much less than the natural linewidth of the transitions. During the experiment, the intensities of these fields are usually an order of magnitude larger than the saturation intensities. We identify power broadening by the 233-nm field and the resulting decrease in the number of absorption depths at both ω_e and ω_h as the primary saturation mechanism in this experiment. Close agreement between numerical simulation and experimental results is obtained if the small-signal absorption coefficients α_e and α_h are replaced by their power-broadened values. Saturation is most evident in the behavior of the conversion efficiency from $\omega_e \rightarrow \omega_h$; small-signal efficiencies [$\Omega_e(0) < 0.005\ \mathrm{cm}^{-1}$] exceed 30% but drop sharply to between 5% and 10% when $\Omega_e(0)$ is raised to 0.02 cm^{-1} because the effective opacity is progressively reduced to the point where the matching condition is no longer attained.

At the higher NL condition, we achieved approximately 1% overall energy conversion efficiency. A VUV pulse energy of 1 μJ was generated using 10 μJ of 233 nm and 50 μJ each of 283 and 406 nm.

In summary, we have demonstrated efficient nonlinear frequency conversion in a quadruply resonant double-Λ atomic system, using collimated laser fields with modest power densities. This work provides the first experimental realization of nonlinear optics in a density-invariant, Rabi-frequency matching regime. Three resonant, arbitrarily phased fields are applied at $z = 0$, and after a characteristic distance, the atom renders itself transparent to all fields through generation of a properly phased, fourth resonant field. Transparency is achieved once the complex ratio of Rabi frequencies in each Λ-channel is equal, and once transparent, no further changes to relative phase or amplitude occur. The overall conversion efficiencies of these all-resonant frequency converters are limited by the preparation energy requirement necessary to establish EIT in the first Λ-channel and by power broadening of the second Λ-channel. We note that the use of a single-isotope atomic medium minimizes the required values of Ω_c [14] but is otherwise unnecessary.

The authors thank H. Xia for experimental assistance and J. Reader and R. Englemann at Opthos Instruments for construction and filling of the custom sidearm cells. This work was supported by the U.S. Air Force Office of Scientific Research, the U.S. Army Research Office, and the U.S. Office of Naval Research.

Note added in proof.—A comprehensive theoretical study of nonlinear optics in double-Λ atomic systems, with emphasis on pulse-matching properties, has recently been published by Korsunsky *et al.* [15].

[1] M. Jain, H. Xia, G. Y. Yin, A. J. Merriam, and S. E. Harris, Phys. Rev. Lett. **77**, 4326 (1996); M. Jain, J. E. Field, and G. Y. Yin, Opt. Lett. **18**, 998 (1993).

[2] A. J. Merriam, S. J. Sharpe, H. Xia, D. Manuszak, G. Y. Yin, and S. E. Harris, Opt. Lett. **24**, 625 (1999); A. J. Merriam, S. J. Sharpe, H. Xia, D. Manuszak, G. Y. Yin, and S. E. Harris, IEEE J. Sel. Top. Quantum Electron. **5**, 1502 (1999).

[3] K. Hakuta, M. Suzuki, M. Katsuragawa, and J. Z. Li, Phys. Rev. Lett. **79**, 209 (1997).

[4] See, e.g., E. Arimondo, in *Progress in Optics*, edited by E. Wolf (North-Holland, Amsterdam, 1996), Vol. XXXV, and references therein.

[5] S. Y. Zhu, M. O. Scully, H. Fearn, and L. M. Narducci, Z. Phys. D **22**, 483 (1992); M. Fleischhauer and M. O. Scully, Opt. Commun. **105**, 79 (1994); O. Kocharovskara and P. Mandel, Phys. Rev. A **42**, 523 (1990).

[6] Experimental work is described by B. Lu, W. H. Burkett, and M. Xiao, Opt. Lett. **23**, 804 (1998); S. Babin, U. Hinze, E. Tinemann, and B. Wellehehausen, Opt. Lett. **21**, 1186 (1996); P. R. Hemmer *et al.*, Opt. Lett. **20**, 982 (1995); A. S. Zibrov, M. D. Lukin, and M. O. Scully, Phys. Rev. Lett. **83**, 4049 (1999). A theoretical study was undertaken by M. D. Lukin, P. R. Hemmer, M. Loeffler, and M. O. Scully, Phys. Rev. Lett. **81**, 2675 (1998).

[7] S. E. Harris, Phys. Rev. Lett. **70**, 552 (1993).

[8] See, e.g., A. V. Smith, W. J. Alford, and G. R. Hadley, J. Opt. Soc. Am. B **5**, 1503 (1988), and references therein.

[9] A. Kasapi, M. Jain, and G. Y. Yin, Appl. Phys. Lett. **35**, 1999 (1996); A. J. Merriam and G. Y. Yin, Opt. Lett. **23**, 1034 (1998).

[10] The fluorescence quantum efficiency is the same at 233 and 186 nm; see, e.g., J. A. R. Samson, *Techniques of Vacuum Ultraviolet Spectroscopy* (Pied Publications, Lincoln, Nebraska, 1967).

[11] A. Kasapi, M. Jain, G. Y. Yin, and S. E. Harris, Phys. Rev. Lett. **74**, 2447 (1995); M. Jain, A. J. Merriam, A. Kasapi, G. Y. Yin, and S. E. Harris, Phys. Rev. Lett. **75**, 4385 (1995).

[12] S. E. Harris and Z. F. Luo, Phys. Rev. A **52**, R928 (1995).

[13] S. Wielandy and A. L. Gaeta, Phys. Rev. A **58**, 2500 (1998).

[14] H. Xia, A. J. Merriam, S. J. Sharpe, G. Y. Yin, and S. E. Harris, Phys. Rev. A **59**, R3190 (1999).

[15] E. A. Korsunsky and D. V. Kosachiov, Phys. Rev. A **60**, 4996 (1999).

Raman Generation by Phased and Antiphased Molecular States

A. V. Sokolov, D. R. Walker, D. D. Yavuz, G. Y. Yin, and S. E. Harris
Edward L. Ginzton Laboratory, Stanford University, Stanford, California 94305
(Received 14 December 1999)

We use molecular deuterium and two driving lasers to demonstrate collinear generation of mutually coherent equidistant sidebands, covering 50 000 cm^{-1} of spectral bandwidth and ranging from 2.94 μm to 195 nm in wavelength. The essential idea is the adiabatic preparation of a single, highly coherent ($|\rho_{ab}| = 0.33$) molecular eigenstate.

PACS numbers: 42.50.Gy, 32.80.Qk, 42.60.Fc, 42.65.Dr

It has recently been predicted that coherent molecular oscillations can produce laser frequency modulation (FM), with a total bandwidth extending over the infrared, visible, and ultraviolet spectral regions, and with a possibility of subfemtosecond pulse compression. The essence of this technique is the use of a Raman transition with a sufficiently large coherence that the generation length and the phase-slip length are of the same order. This coherence is established by driving the molecular transition with two single-mode laser fields, slightly detuned from the Raman resonance so as to excite a single molecular eigenstate. Molecular motion, either in phase with the driving force (Raman detuning below resonance) or antiphased (detuning above resonance), in turn modulates the driving laser frequencies, causing the collinear generation of a very broad FM-like spectrum [1].

This Letter describes the first experimental results for this technique. Using molecular deuterium (D_2), we demonstrate collinear generation of a Raman spectrum extending over 50 000 cm^{-1}. The spectrum consists of seventeen mutually coherent sidebands, spaced by the fundamental vibrational frequency of D_2 (2994 cm^{-1}), and ranging in wavelength from 2.94 μm to 195 nm.

There is prior work on broadband coherent Raman generation. Several groups have studied the enhancement of stimulated Raman scattering by application of the first Stokes component collinearly with the pump laser beam; they have demonstrated increased conversion efficiency, improved reproducibility, and small divergence of the anti-Stokes beams [2–4]. Hakuta *et al.* have demonstrated collinear Raman sideband generation in solid molecular hydrogen (H_2) [5]. Nazarkin *et al.* have shown efficient laser frequency modulation by impulsively exciting a coherent vibration of SF_6 [6]. In other experiments, Ruhman *et al.* have used impulsive stimulated Raman scattering to observe coherent molecular vibrations in the time domain [7]. The phased and antiphased excitation techniques, and the resultant maximum coherence of a molecular state, have not been discussed.

Our experimental setup is shown in Fig. 1. We use two transform-limited laser pulses at wavelengths of 1.0645 μm and 807.22 nm, such that the (tunable) laser-frequency difference is approximately equal to the fundamental vibrational frequency in D_2. The first laser is a Quanta-Ray GCR-290 Q-switched injection-seeded Nd:YAG laser. Its output is attenuated to produce 100 mJ, 12 ns transform-limited pulses at a 10 Hz repetition rate. The laser linewidth is calculated to be $\delta\omega_{\text{laser}} = 37$ MHz. The second laser is a lab-built Ti:sapphire system, injection seeded from an external-cavity laser diode and pumped by the second harmonic of a separate Q-switched Nd:YAG laser. This laser produces 75 mJ, 16 ns transform-limited pulses at the seeding laser wavelength. This wavelength can be tuned precisely and is monitored by a Burleigh WA-1500 Wavemeter (resolution of 50 MHz). The two driving laser pulses are synchronized by adjusting the delay between the two Nd:YAG laser Q-switch trigger pulses. The laser beams are combined on a dichroic beam splitter and are loosely focused to a nearly diffraction-limited spot in a D_2 cell. The 1.06 μm laser spot size is 460 μm, and the 807 nm laser spot size is 395 μm.

We determine the Raman resonance by scanning the Ti:sapphire laser frequency and measuring weak Stokes gain at a low pump intensity and a small D_2 density. The D_2 cell is cooled by liquid nitrogen to $T = 77$ K; the length of the cooled region is 50 cm. Cooling reduces the Doppler linewidth to 260 MHz and increases the population of the ground rotational state of D_2 to 60%.

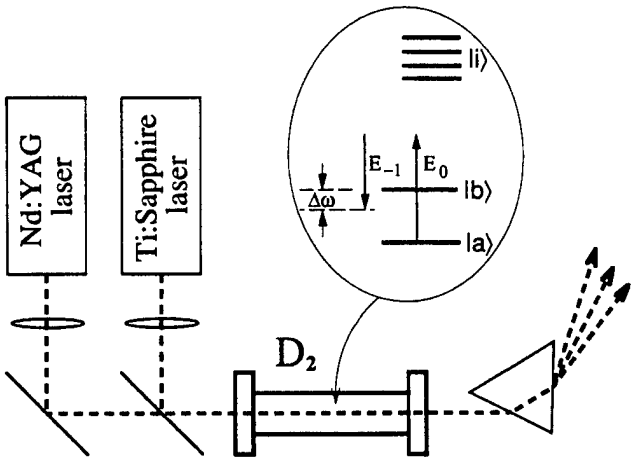


FIG. 1. Experimental setup and energy level diagram for coherent molecular excitation and collinear Raman generation. The Raman detuning $\Delta\omega$ (positive as shown) is set by the driving laser frequencies.

 0031-9007/00/85(3)/562(4)$15.00

The collisional dephasing linewidth in D_2 at $T = 77$ K is 450 kHz/torr, as deduced from room-temperature data [8].

When we tune the driving infrared lasers to within 1 GHz from the Raman resonance, we see a bright beam of white light at the output of the D_2 cell. We disperse the spectrum with a prism and project (without collimating or focusing) onto a white scintillating screen (60 cm wide, 270 cm away from the cell). At a D_2 pressure of 50–100 torr and at a Raman detuning of a few hundred MHz, we observe up to thirteen anti-Stokes sidebands and two Stokes sidebands, in addition to the two driving frequencies. These sidebands are spaced by 2994 cm^{-1} and range from 2.94 μm to 195 nm in wavelength. The next Stokes sideband has a wavelength of 24 μm and is difficult to detect. The next (fourteenth) anti-Stokes sideband has a wavelength of 184 nm and is absorbed by air.

Figure 2 shows pictures of the spectrum taken with a digital color camera with a single-shot exposure at a fixed aperture size. Starting from the left, the first two sidebands are the driving frequencies, and the next four are anti-Stokes sidebands in real color (red, green, blue, and violet); beginning at the fifth anti-Stokes, the sidebands are in the ultraviolet and only fluorescence is visible. Figures 2(a)–2(c) show the spectrum generated at a D_2 pressure of $P = 71$ torr and a Raman detuning of $\Delta\omega = -400 \pm 25$ MHz in part (a), $\Delta\omega = 100 \pm 25$ MHz in part (b), and $\Delta\omega = 700 \pm 25$ MHz in part (c). The smooth near-Gaussian beam profiles for nearly all sidebands, as shown in Figs. 2(a)–2(c), demonstrate collinear anti-Stokes generation in a regime of high molecular coherence. At higher pressures the generation is no longer collinear and the anti-Stokes sidebands emerge in circles of increasing diameter. An example at a pressure of 350 torr and $\Delta\omega = 700$ MHz is shown in Fig. 2(d).

Figure 3 shows Stokes and anti-Stokes energy spectra for three different values of the Raman detuning and contrasts the on-resonance generation at $P = 72$ torr (triangles and solid line) with the generation below resonance (circles and dashed line) and above resonance (squares and dotted line). We observe that on-resonance generation is less efficient than off-resonance generation for all of the anti-Stokes sidebands, and that generation below resonance is more efficient than above resonance. Note that we generate hundreds of μJ per pulse (at a 10 Hz repetition rate) at sidebands far into the UV.

We now proceed with the discussion of these results. We consider a set of equidistant Raman sidebands with complex field envelopes E_q and carrier frequencies ω_q, tuned close to the Raman transition $|a\rangle \rightarrow |b\rangle$, and far detuned from the upper electronic states $|i\rangle$. Propagation of each sideband is governed by the slowly varying envelope equation in local time [1]:

$$\frac{\partial E_q}{\partial z} = -j\eta\hbar\omega_q N(a_q\rho_{aa}E_q + d_q\rho_{bb}E_q + b_q^*\rho_{ab}E_{q-1} + b_{q+1}\rho_{ab}^*E_{q+1}), \quad (1)$$

where ρ_{aa} and ρ_{bb} are populations of states $|a\rangle$ and $|b\rangle$, ρ_{ab} is the Raman coherence, N is the molecular density, and $\eta = (\mu/\varepsilon_0)^{1/2}$. Expressions for the dispersion and coupling constants a_q, b_q, and d_q are given in Ref. [1].

When the magnitude of the coherence ρ_{ab} approaches its maximum value of 0.5, the coupling terms in Eq. (1) (last two terms) are of the same order as the dispersion terms (first two terms). In such a high-coherence regime, the generation length becomes comparable to the phase-slip length, and efficient generation proceeds collinearly, with phase matching playing a small role [9]. To the extent that dispersion can be neglected completely, theory predicts a pure sinusoidal frequency modulation of a laser beam, as a result of its propagation through the coherent medium [1].

The excitation of the molecular states $|a\rangle$ and $|b\rangle$ is described by an effective two-by-two Hamiltonian [1]:

$$H_{\text{eff}} = -\frac{\hbar}{2}\begin{bmatrix} A & B \\ B^* & D - 2\Delta\omega \end{bmatrix}, \quad (2)$$

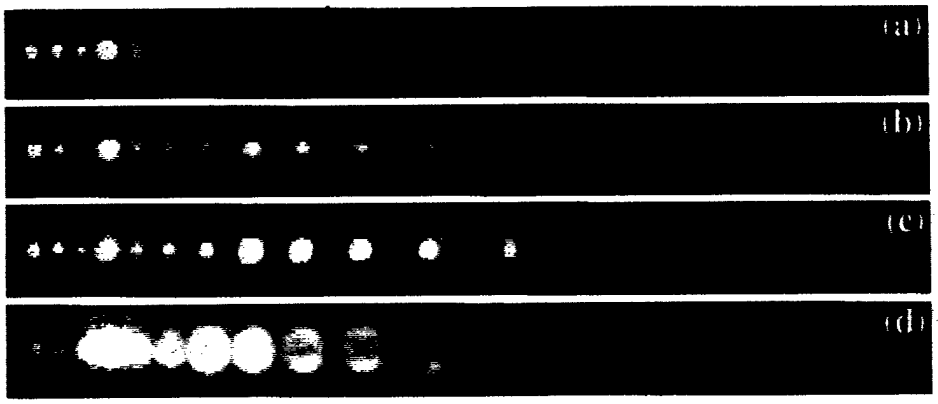


FIG. 2 (color). Spectrum generated in the setup of Fig. 1 at (a) $P = 71$ torr and $\Delta\omega = -400$ MHz, (b) $P = 71$ torr and $\Delta\omega = 100$ MHz, (c) $P = 71$ torr and $\Delta\omega = 700$ MHz, and (d) $P = 350$ torr and $\Delta\omega = 700$ MHz. We observe the two driving infrared fields (on the left), and multiple generated visible and ultraviolet anti-Stokes sidebands. To reduce camera saturation for (a)–(c), the first four anti-Stokes beams are attenuated by a factor of 100; the driving field at 807 nm and also the fifth anti-Stokes beam are attenuated by 10. In (d), the 807 nm beam and the first four anti-Stokes beams are attenuated by 10.

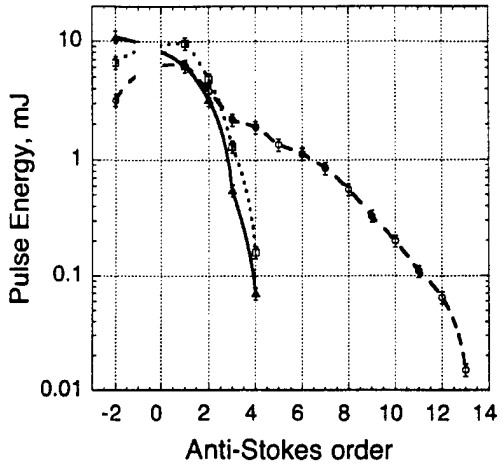


FIG. 3. Pulse energies generated in the setup of Fig. 1 at $P = 72$ torr. The triangles show on-resonance generation ($\Delta\omega = 0$), the circles show generation by phased ($\Delta\omega = 500$ MHz) and the squares by anti-phased ($\Delta\omega = -200$ MHz) states of D_2.

where $A = \sum_q a_q |E_q|^2$, $B = \sum_q b_q E_q E_{q-1}^*$, and $D = \sum_q d_q |E_q|^2$. The Raman detuning $\Delta\omega$ is the difference between the molecular transition frequency $\omega_b - \omega_a$ and the frequency of the driving force $\omega_q - \omega_{q-1}$. The collisional dephasing is included in the equation for the off-diagonal density-matrix element (the coherence) ρ_{ab}. For the preparation of maximum coherence it is essential that the dephasing time is longer than the laser pulse length (as is the case in our experiment).

The molecular eigenstate which evolves from the ground state as the field amplitudes are increased and the coherence which is obtained from it are

$$|+\rangle = \cos\frac{\theta}{2}\exp\left(j\frac{\varphi}{2}\right)|a\rangle + \sin\frac{\theta}{2}\exp\left(-j\frac{\varphi}{2}\right)|b\rangle,$$
$$\rho_{ab} = \frac{1}{2}\sin\theta\exp(j\varphi), \tag{3}$$

where $B = |B|\exp(j\varphi)$ and $\tan\theta = 2|B|/(2\Delta\omega - D + A)$. When the Raman detuning is larger than the driving laser linewidth, the molecular system follows this eigenstate adiabatically, with the sign of ρ_{ab} determined by the sign of the Raman detuning. In a resonant, nonadiabatic regime the molecules exhibit two-photon Rabi flopping at a frequency B.

In our experiment the Doppler broadened linewidth (260 MHz) of the 2994 cm^{-1} transition of D_2 is larger than both the collisional dephasing rate (32 MHz for $P = 72$ torr) and the laser linewidth (37 MHz). When we tune close to the center of the Doppler line, we expect adiabatic preparation for most of the molecules in both wings of the velocity distribution, with significant magnitude of their excitation when the coupling parameter $B > \delta\omega_{\text{laser}}$. The phases of these excitations are positive or negative depending on the sign of the Raman detuning, such that the contributions of these molecules partially cancel each other, resulting in a smaller integrated value of the coherence ρ_{ab} at the center frequency. However, when we detune to either side of the Doppler line, we expect adiabatic preparation of nearly all of the molecules in the same (phase or antiphased) state [Eqs. (3)], resulting in a larger total coherence and more efficient Raman generation. Similar behavior has been observed in saturated coherent anti-Stokes Raman spectroscopy [10].

Figure 4 illustrates this behavior. Parts (a) and (b) show the first anti-Stokes pulse energy as a function of the Raman detuning, both at low pressure ($P = 6$ torr). Here, the depletion of the applied fields is not important, and the generated energy is proportional to the square of the Doppler-averaged coherence [Eq. (1)]. In Fig. 4(a), the 1.06 μm laser pulse energy is 5.4 mJ and $B = 39$ MHz $\approx \delta\omega_{\text{laser}}$; for these conditions the anti-Stokes generation peaks on resonance. For Fig. 4(b) we apply full power of the driving lasers, so that $B = 168$ MHz $\gg \delta\omega_{\text{laser}}$, and observe that the anti-Stokes generation peaks on both sides of the resonance, with a dip exactly on resonance. The peak at positive $\Delta\omega$ is higher, because, as the laser power increases as a function of time, this peak Stark-shifts toward resonance. We solve the full density-matrix equations numerically for the experimental driving laser pulses, calculate the (complex) coherence ρ_{ab} at the pulse peak for different values of Raman detuning, and average it over the Doppler distribution. The solid lines in Figs. 4(a) and 4(b) show the square of the Doppler-averaged coherence (no scaling). We calculate that the maximal value of the coherence that we achieve in our experiment is $|\langle\rho_{ab}\rangle_{\text{Doppler}}| = 0.33$.

Figure 4(c) shows qualitatively similar behavior at a higher D_2 pressure ($P = 72$ torr). The filled circles represent the sum of all generated Stokes and anti-Stokes sidebands. The open circles and triangles show the first and second anti-Stokes sidebands. All measurements for Figs. 3 and 4 are done with a Molectron J4-09 pyroelectric detector. Each point in the plots is an average over eight laser shots, with error bars showing the standard deviation for each data set.

In order to investigate adiabatic and nonadiabatic molecular excitation we measure the total laser energy transmitted through the D_2 cell (including all applied and generated sidebands) as a function of the Raman detuning. When the molecular evolution is adiabatic, we expect all molecules to return to the ground state as the laser fields decrease at the falling edge of the applied pulses. We observe a dip in transmission on Raman resonance, confirming that energy is left in the molecular system due to nonadiabatic behavior. For comparison we numerically calculate the probability for the molecular system to remain in the adiabatic eigenstate $|+\rangle$ [Eqs. (3)], averaged over the Doppler distribution [solid line in Fig. 4(d)], again demonstrating good qualitative agreement between theory and experiment. By comparing Figs. 4(c) and 4(d), we note that the Raman generation peaks at detunings where the molecular evolution is completely adiabatic.

We note the connection of this work to the coherent population transfer [11] and to electromagnetically induced

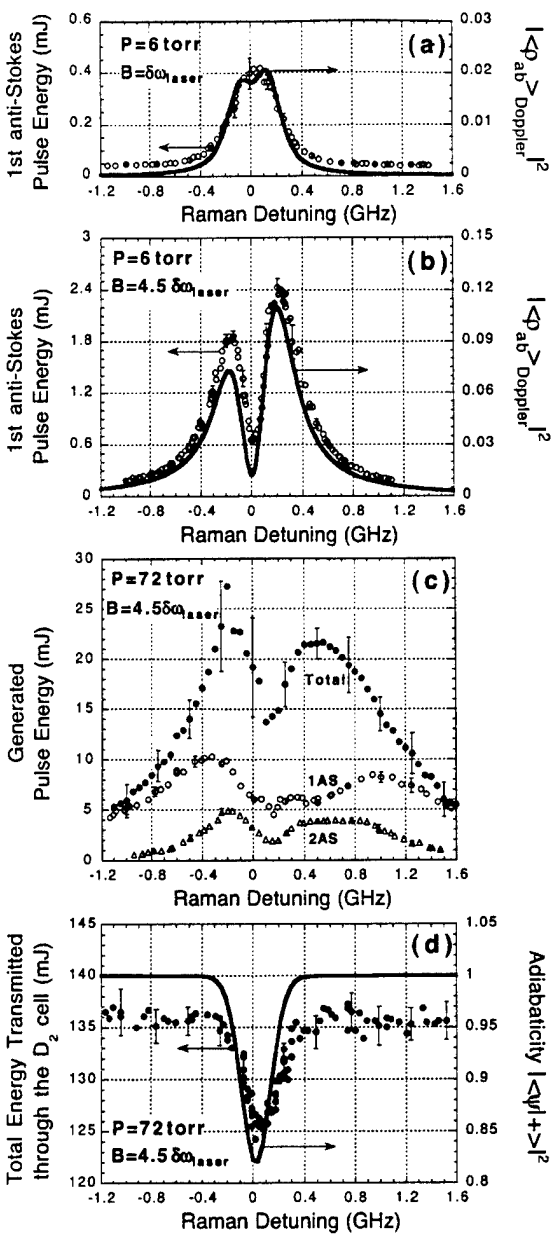


FIG. 4. Raman generation by adiabatic eigenstates in D_2. (a) First anti-Stokes generation by weakly driven vibrations ($B \approx \delta\omega_{\text{laser}}$) at $P = 6$ torr. (b) First anti-Stokes generation by strongly driven vibrations ($B \gg \delta\omega_{\text{laser}}$) at $P = 6$ torr. In both (a) and (b) the solid lines show the calculated square of the Doppler-averaged coherence ρ_{ab} at the pulse peak. (c) Sum over pulse energies of all generated Raman sidebands (filled circles), first anti-Stokes generation (open circles), and second anti-Stokes generation (triangles) for $B \gg \delta\omega_{\text{laser}}$ and $P =$ 72 torr. (d) Total energy in all applied and generated sidebands at the output of the D_2 cell. The solid line represents the calculated fraction of the molecular population in the eigenstate which evolves adiabatically from the ground state [Eq. (3)].

transparency (EIT) [12]: Each involves the preparation of the adiabatically evolving eigenstate. In three-state EIT this is done by using quantum interference or optical pumping. Here it is done by fixed or adiabatically changing detuning. Both EIT and this process require that the number of photons in the laser pulse exceed the number of molecules in the laser path [12,13]. Both allow nonlinear optics at maximum coherence, where the role of phase matching is greatly reduced [9]. But, differing from EIT, here, the refractive indices at the various sidebands are reduced (in the antiphased case) [1] but are not equal to unity.

In summary, this Letter demonstrates broadband collinear Raman generation by adiabatically prepared phased and antiphased states in molecular deuterium. We show that, in agreement with theory, generation maximizes at a finite detuning on either side of the Raman resonance. We believe it will be possible to recombine the generated sidebands and to use spectral modification techniques [14] to synthesize specified subfemtosecond time structures in a target cell.

This work was supported by the U.S. Army Research Office, the U.S. Air Force Office of Scientific Research, and the U.S. Office of Naval Research. D. R. W. also acknowledges support from the Fannie and John Hertz Foundation.

[1] S. E. Harris and A. V. Sokolov, Phys. Rev. A **55**, R4019 (1997); S. E. Harris and A. V. Sokolov, Phys. Rev. Lett. **81**, 2894 (1998); A. V. Sokolov, D. D. Yavuz, and S. E. Harris, Opt. Lett. **24**, 557 (1999); A. V. Sokolov, Opt. Lett. **24**, 1248 (1999).

[2] V. Schulz-von der Gathen, T. Bornemann, V. Kornas, and H. F. Dobele, IEEE J. Quantum Electron. **26**, 739 (1990).

[3] L. L. Losev and A. P. Lutsenko, Quantum Electron. **23**, 919 (1993); G. S. McDonald, G. H. C. New, L. L. Losev, A. P. Lutsenko, and M. Shaw, Opt. Lett. **19**, 1400 (1994).

[4] H. Kawano, Y. Hirakawa, and T. Imasaka, IEEE J. Quantum Electron. **34**, 260 (1998).

[5] K. Hakuta, M. Suzuki, M. Katsuragawa, and J. Z. Li, Phys. Rev. Lett. **79**, 209 (1997).

[6] A. Nazarkin, G. Korn, M. Wittman, and T. Elsaesser, Phys. Rev. Lett. **83**, 2560 (1999).

[7] S. Ruhman, A. G. Joly, and K. A. Nelson, IEEE J. Quantum Electron. **24**, 460 (1988).

[8] D. A. Russel and W. B. Roh, J. Mol. Spectrosc. **124**, 240 (1987).

[9] M. Jain, H. Xia, G. Y. Yin, A. J. Merriam, and S. E. Harris, Phys. Rev. Lett. **77**, 4326 (1996); A. J. Merriam, S. J. Sharpe, H. Xia, D. Manuszak, G. Y. Yin, and S. E. Harris, Opt. Lett. **24**, 625 (1999).

[10] R. P. Lucht and R. L. Farrow, J. Opt. Soc. Am. B **5**, 1243 (1988).

[11] K. Bergmann, H. Theuer, and B. W. Shore, Rev. Mod. Phys. **70**, 1003 (1998).

[12] S. E. Harris, Phys. Today **50**, No. 7, 36 (1997); M. O. Scully and M. S. Zubairy, *Quantum Optics* (Cambridge University Press, Cambridge, England, 1997).

[13] D. D. Yavuz, A. V. Sokolov, and S. E. Harris, Phys. Rev. Lett. **84**, 75 (2000).

[14] C. W. Hillegas, J. X. Tull, D. Goswami, D. Strickland, and W. S. Warren, Opt. Lett. **19**, 737 (1994); A. M. Weiner, Prog. Quantum Electron. **19**, 161 (1995); T. Baumert, T. Brixner, V. Seyfried, M. Strehle, and G. Gerber, Appl. Phys. B **65**, 779 (1997).